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# Determination and quantification of NORM radionuclides

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## Abstract

This paper discusses methodologies which can be used to identify naturally occurring radionuclides in substances using automatic spectrum analysis. The possibility to use automatic spectrum analysis in order to obtain an estimation of the activity concentration of the detected radionuclides is examined. The goal is to determine whether or not the activity concentration of a measured substance meets the regulation defined in the new upcoming European Basic Safety Standards with respect to naturally occurring radioactive materials (NORM). Existing techniques which are primarily used in lab settings are applied in industrial settings and tested. These techniques include artificial neural networks and automated spectrum analysis. The methodology is assessed and a comparison is made between NaI(Tl) and LaBr(Ce) based multi-channel analysers. The methodology is applied to several NORM materials with distinct activity concentrations: zirconium, ilmenite, bauxite, and fluorspar. In this way a methodology is constructed which can be applied in situ to allow determination and quantification of naturally occurring radionuclides.

## Introduction

Radionuclides of the natural radioactive series of uranium and thorium are present everywhere in the earth crust. Concentration of these nuclides is depending on the composition of the soil. Besides uranium and thorium nuclides, an amount of potassium-40 is also present. All these nuclides produce radiation doses to all human beings. In addition, industrial processes can lead to an accumulation of naturally occurring radioactive materials (NORM) due to operations in end-, by- or waste-products. The concentration of nuclides can be accumulated in such a way that radiation protection is necessary.

A uniform approach towards NORM by all European member states is a primary goal. The European Commission is therefore currently recasting Council Directives with respect to natural radiation sources (European Commission 2010). One of the new elements is the construction of a positive list of industrial activities in the non-nuclear

sector that may be subject to notification. As already described in UNSCEAR 2000 and ICRP Publication 75 the acceptable dose rate threshold can be aligned with the activity concentration of materials. As a result the activity concentration will be used to determine whether regulatory authorities need a prior authorisation.

This means it is important to have a technique to determine the activity concentration of NORM. Since the activity concentration has to be measured in all the products, by-products and residues in an industrial process, a fast and usable methodology is mandatory. Nowadays the activity concentration is generally being measured by sample analysis which is a cumbersome, time and money consuming job.

In this work the objective is to construct a tool that can automatically decide in an industrial setting whether a measured substance complies with the new European Directive's activity concentration limits. This has to occur in known geometries by using a relatively cheap NaI(Tl) probe-based multichannel analyzer (MCA) connected to a tablet-PC. Custom software on the tablet steers the MCA to capture a spectrum and to interpret the spectrum straightaway. Based on the spectrum, the weight of the substance and its density, an estimation of the activity concentration is made and presented to the user.

To assess the technique proposed we captured several spectra of distinct NORM materials in a known geometry of a NORM handling company. The company stores and handles sugars, fertilizers, chemicals, minerals, iron, steel and wood products. The handling comprises bagging, repacking, sieving, sifting, weighing, mixing and conditioned storage in contamination-free warehouses. The accuracy of the tool was assessed by comparing the results with the results of an accepted method based on sample analysis.

In this work we focus on tool support. The objective is to construct a tool that can automatically decide in an industrial setting whether a measured substance complies with the new European Directive's activity concentration limits.

## Tool support for determination and quantification of NORM radionuclides in industrial settings

An important goal of our current research project is to make an inventory of the issues of NORM in a NORM handling company in collaboration with the Federal Agency of Nuclear Control.

The measurement tool was designed in such a way that at the end of the project the employees of the industry can use it independently. Therefore it has to be user-friendly and applicable in distinct industrial circumstances. Software is constructed that can be easily adapted according to the environmental context: several distinct geometries are present so that the end user can select the geometry that is applicable to the current context-of-use. For example: when the substance is contained in a shipping container, this geometry can be selected and subsequently parameters with respect to a shipping container (dimensions, probe location, container wall properties etc.) can be filled out by the user. On the other hand when the substance is contained in a bigbag (common used bag in industry containing dry bulk substances) this can be selected by the user and geometry properties can be filled out by the user accordingly (diameter, circumference, etc.).

As big bags were the most common geometry in this company, the tool was at first adjusted for this model (Fig. 1). Big bags contain dry bulk substances with a mass typically between 1 and 2 ton. Activity concentrations of nuclides in big bags were determined on the side but also on the top of the bag. The measurements on the top correspond the most to the sample analysis.

In practice, the bags were placed outside the warehouse to avoid interference with radiation caused by other bags. Geometry of the bag was determined (height, net mass, perimeter, volume). Two probes (NaI(Tl) and LaBr(Ce)) were set on the top and in contact with the bags and were connected with the spectrometer and software. Measurements were repeated 10 times for 15 minutes in order to control the reproducibility of measurements. A spectrum was automatically generated and analysed.

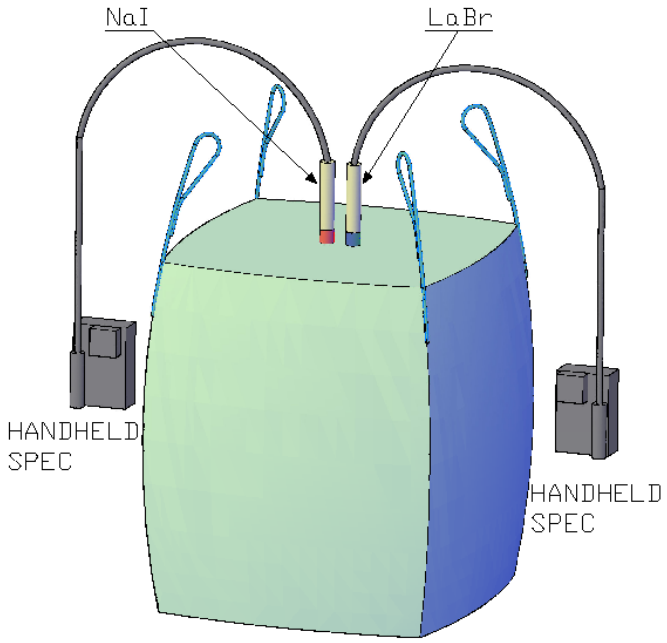


**Fig. 1. A bigbag with two multi-channel analysers (NaI(Tl) and LaBr(Ce)) recording a spectrum of a known substance.**

In the following sections two techniques are discussed which can be applied to identify NORM radionuclides by using Artificial Neural Networks and to quantify the activity concentration of NORM radionuclides by applying energy calibration by modelling the measured container. Both techniques make use of automated spectrum analysis of the spectra recorded in the industrial setting of the NORM handling company. The accuracy of the tool was checked by comparison of the analyses with sample analysis.

## Determination of nuclides with Artificial Neural Networks (ANN)

This section elaborates on a technique to classify NORM radionuclides using artificial neural networks. The following section will discuss a technique to estimate the activity concentration.

## Artificial Neural Networks for gamma-ray spectrum analysis

Artificial Neural Networks ANN simulate biological neural networks in a computational model that can be implemented in a software system. ANN already have a long history of applications in physics and chemistry in general and in nuclear science in particular with proven successful results on spectrum analysis (Long 1997) (Keller et al. 1994) (Yoshida et al. 2002) (Saritha et al. 2009). A key characteristic of ANN is the excellent performance on coping with data containing noise. Milford identified the generalization ability of ANN as the reason for this performance when increasing amounts of Gaussian noise were added to spectra (Milford 2002). Saritha et al. And Keller et al. compared a neural network to an Optimal Linear Associative Memory (OLAM) for the classification of linearly separable data (Saritha et al. 2009) and (Keller et al. 1994). While OLAM was superior to a neural network when Monte Carlo generated spectra were used, the ANN was again found to be the best solution for noisy data.

## Practical Approach

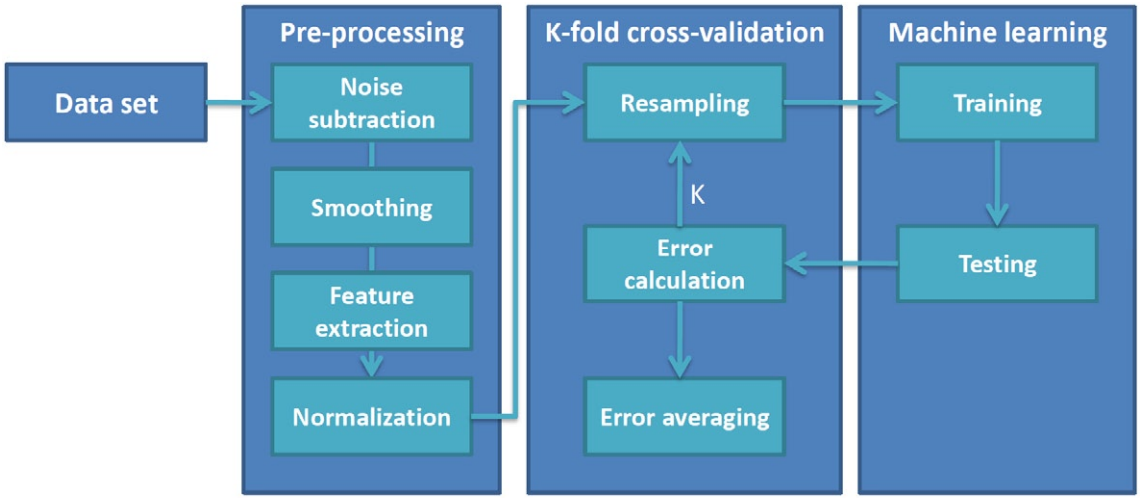


**Fig. 2. Schematic overview of the machine learning sequence.**

Figure 2 presents a schematic overview of the machine learning sequence of the ANN that has been implemented. The input data set is a collection of raw spectra obtained by the measurements in the industrial case study. The machine learning sequence consists of three steps: pre-processing, cross-validation, and machine learning.

Before the spectral data can be fed to the ANN a pre-processing step is applied to extract the most relevant data from the spectrum. Although this is strictly not necessary since the ANN can learn which data are irrelevant. Feeding 512 to 4096 channels used by an MCA to the ANN would be inefficient and decrease the training speed. Using trial and error the following pre-processing techniques generated the best results on the spectra recorded in the industrial case study: noise subtraction, running-average smoothing, peak-extraction based on Milford, and normalization (Milford 2002). The peak extraction algorithm investigates a specific number of channels surrounding a

point to determine whether the point is a local maximum and thus a peak. This reduces the complexity of the input layer.

Figure 3 presents the implemented ANN. The ANN is a fully interconnected feed-forward network with one hidden layer. Back-propagation learning is used to train the ANN. Because of the pre-processing step, the number of input neurons is reduced to twelve, i.e. the respective position and height of six key peaks that can identify the presence of a radionuclide in the spectrum. The number of neurons in the hidden layer has to be chosen carefully in order to find a balance between the model's complexity and the generalization capacity of the ANN.

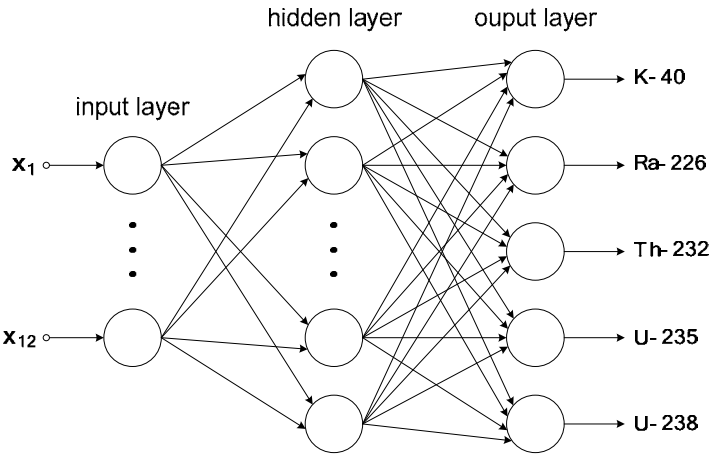


**Fig. 3. Model of the implemented ANN.**

To validate the ANN k-fold cross-validation was applied with k=10. This means the input data set was divided in ten equal collections of spectra from which nine are used to train the ANN and the remaining one is used for validation. Afterwards the error is calculated and the input data set is resampled for another run. This is repeated until the cross-validation error < 1%.

## Results

The gamma spectra are recorded using a NaI(Tl) and LaBr(Ce) detector in the industrial environment. A training set of eighty spectral samples was used that represented all detectable isotopes (K-40, Ra-226, Th-232, U-235, and U-238) in equal numbers. The k-fold cross-validation was applied to exploit the reasonably small data set of eighty samples. A series of ten repeated 10-fold cross-validations were used to verify the performance of the ANN.

In order to optimize the performance of the network, the following parameters were isolated and compared by their accuracy, mean-squared error (MSE) and mean-absolute error (MAE):

- Momentum: scaling factor to specify to what degree the weights of the neurons change from the previous step of the learning algorithm. The momentum appeared to be acceptable with respect to the accuracy and the MSE between 0.1 and 0.3.
- Hidden-layer size: the MSE and MAE both reach a minimum when the hidden layer consists of four neurons.

Furthermore the learning rate and the number of epochs (iterations of an entire training set) were taken into account.

The results showed that the ANN classified the spectra with an accuracy of 85% on average which corresponds to an error rate of 15%. This large error was due to the problems that the ANN had with the classification of U-238. The best performance of the ANN on U-238 was 87%. The other isotopes (K-40, Ra-226, Th-232, and U-235) were classified with an accuracy of 100%.

## Quantification of NORM radionuclides

In earlier work we discussed how the activity concentration of NORM radionuclides was quantified using a NaI(Tl)-based multi-channel analyser (MCA) using spectrum analyses on spectra captured in an industrial setting (Pellens et al. 2010). In this section we will compare results of spectrum analysis captured by an MCA equipped with a NaI(Tl) probe with spectra from the same substances captured in the same environmental circumstances but with an MCA equipped with a LaBr(Ce) probe. Furthermore the technique is assessed by comparing these results to the results of sample analysis performed by a certified institution.

### Practical approach

The approach consists of automated spectrum analyses performed by customized software. Four substances known as NORM were measured in bigbags containing between 1000 kg and 2000 kg. The software was adapted to cope with spectra recorded by either NaI(Tl) and LaBr(Ce) probes. The final output of the software was an estimation of the activity concentration of NORM-radionuclides present in the measured substance. Measurements taken with NaI(Tl) and LaBr(Ce) are compared to results from sample analyses taken from the respective bigbags. The analyses on 250 g samples were performed by a certified lab. In the next section this comparison will be discussed.

The automated spectrum analysis was applied to ten spectra recorded on each substance. The same automated analysis algorithm was applied to the distinct substances.

Up to now, four distinct substances were considered: zirconium, ilmenite, bauxite, and fluorspar. The substances were chosen in a way that there was a variety in the respective activity concentration of the radionuclides present.

### Results

The results are presented in three tables. Each table presents the results of one NORM radionuclide by comparing the automated spectrum analysis of spectra recorded with the two types of probes (Na(Tl) and LaBr(Ce)), the sample analysis by the certified lab and the recommendation as described in the reviewed basic safety standards by the European Commission. The results are presented for each of the four substances considered in this research.

**Table 1. Comparison of spectral analysis results of 900s measurements using a NaI(Tl) and LaBr(Ce) probe with results of a certified lab for the Th-232 radionuclide.**

| Substance | NaI(Tl) | | LaBr(Ce) | | Certified lab SA | EC BSS |
|---|---|---|---|---|---|---|
| | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | |
| Zirconium | 0.53 | 6 | 0.52 | 5 | 0.50 | |
| Ilmenite | 0.28 | -16 | 0.36 | 6 | 0.34 | |
| Bauxite | 0.29 | -2 | 0.33 | 12 | 0.30 | |
| Fluorspar | <MDA = 0.03 | | 0.08 | | <MDA = 0.02 | |

Table 1 presents the results of the Th-232 radionuclide. The sample analysis shows that the performance of the automated sample analysis of the zirconium spectra are nearly the same for the two different probes. There is an overestimation of 5% to 6% in comparison to the certified sample analysis. The activity concentration of Th-232 in ilmenite is performed way better with the LaBr(Ce) probe (6% overestimation) in comparison to the NaI(Tl) (underestimation of 16%). The automated analysis of bauxite presents a different picture: the analysis of the NaI(Tl) probe delivers a better result. Finally the activity concentration of fluorspar does not exceed the minimum detectable activity (MDA) for the NaI(Tl) analysis and the sample analysis. We can conclude that the automated spectrum analysis for the LaBr(Ce) probe delivers an overestimation of the activity concentration of Th-232 in comparison to the NaI(Tl) and sample analysis results. This introduces a more safe result than the NaI(Tl) which may deliver an underestimation. The last column of the table compares the results with the revised basic safety standards. None of the substances exceeds the limit of 1 $Bq \cdot g^{-1}$ for the Th-232 series.

**Table 2. Comparison of spectral analysis results of 900s measurements using a NaI(Tl) and LaBr(Ce) probe with results of a certified lab for the U-238 radionuclide.**

| Substance | NaI(Tl) | | LaBr(Ce) | | Certified lab SA | EC BSS |
|---|---|---|---|---|---|---|
| | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | |
| Zirconium | 1.79 | -8 | 2.13 | 9 | 1.94 | |
| Ilmenite | 0.09 | | <MDA = 0.053 | | <MDA = 0.31 | |
| Bauxite | <MDA = 0.05 | | 0.25 | | <MDA = 0.58 | |
| Fluorspar | 0.08 | | 0.19 | | <MDA = 0.40 | |

Table 2 summarizes the results of the same analyses for the U-238 radionuclide. According to the sample analysis results of the certified lab, only zirconium exceeds the minimum detectable activity. The activity concentration delivered by the automated spectrum analysis of the NaI(Tl) and LaBr(Ce) spectra are comparable: an underestimation of 8% is produced by the NaI(Tl) and an overestimation of 9% by the LaBr(Ce). The activity concentration exceeds the limit of 1 $Bq \cdot g^{-1}$ for the U-238 series.

**Table 3. Comparison of spectral analysis results of 900s measurements using a NaI(Tl) and LaBr(Ce) probe with results of a certified lab for the Ra-226 radionuclide.**

| Substance | NaI(Tl) | | LaBr(Ce) | | Certified lab SA | EC BSS |
|---|---|---|---|---|---|---|
| | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | Rel. Error (%) | Act Conc ($Bq \cdot g^{-1}$) | |
| Zirconium | 1.8 | -15 | 2.1 | -2 | 2.14 | |
| Ilmenite | <MDA = 0.04 | | <MDA = 0.053 | | 0.04 | |
| Bauxite | 0.27 | -8 | 0.27 | -9 | 0.30 | |
| Fluorspar | 0.11 | -37 | 0.19 | 7 | 0.18 | |

Finally table 3 presents the results of the analysis of the Ra-226 radionuclide. For the substances zirconium and fluorspar the LaBr(Ce) outperforms the NaI(Tl): a respective underestimation of 2% and overestimation of 7% versus an underestimation of 15% and 37% for the Ra-226 activity concentration. The results of the spectrum analysis for bauxite are comparable. Again, the only exceeding activity concentration in comparison to the basic safety standards requirements is detected for zirconium.

### Discussion

Looking at the results presented in the previous section, a conclusion can be made that the LaBr(Ce) performs better than the NaI(Tl) when automated spectrum analysis is applied en builds assurance because the error is mostly an overestimation. The results are significantly better when Ra-226 has to be quantified. The comparison of the performance on U-238 is about the same, but only zirconium exceeded the minimum detectable activity. The results of the Th-232 analyses show distinct performances in comparison to the certified sample analysis.

An important issue in the automated spectral analysis seemed to be the detection and quantification of U-238. The certified method using sample analysis could not deliver a justifiable quantification of the activity concentration for three of the four substances. The automated spectral analysis method delivered an estimation of the activity concentration below the minimum detectable activity of the certified lab. Using the automated spectrum analysis the proposed method is able to quantify the activity concentration accurately enough to determine whether the activity concentration is below or above the threshold used in the new European basic safety standards. Further research is required to narrow down the limitations of this methodology to determine when a more accurate analysis is necessary, i.e. when the activity concentration approximates the basic safety standards' limits.

## Conclusions

Automated spectrum analysis was applied on spectra recorded in an industrial setting. The main objective was to study whether automated spectrum analysis can be used in providing a fast tool to check the applicability of the new European basic safety standards with respect to NORM material. A methodology has been constructed consisting of a tablet-PC running a software that steers a measurement and interprets

the spectrum accordingly to approximate the activity concentration of the measured substance.

Both machine learning techniques and traditional automated spectrum analysis were studied in order to determine and quantify NORM radionuclides. Spectra were recorded with two different probes (NaI(Tl) and LaBr(Ce)). Results showed a better performance of the analyses with a LaBr(Ce) probe. In this paper, we showed one can use an ANN to determine the presence of K-40, Ra-226, Th-232 and U-235 with an accuracy of 100% while the determination of the presence of U-238 is 87%. It is in our intention to use these figures to optimize the quantification algorithms used in the automatic spectral analysis.

Results of analyses of four investigated NORM materials were compared with the Council Directives (whether the measured values were in the safe zone, around the threshold, or above the threshold). The four substances which were the subject of this study were accurately classified according to the basic safety standards taking the relative error into account.

Further optimization of the tool will be an important objective of this project. The tool will be able to tell in a very short period if a company needs notification according to the new European basic safety standards.

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The European Commission is currently in the process of recasting five Council Directives: Basic Safety Standards Directive (96/29/Euratom), Medical Directive (97/43/Euratom), Directive on High Activity Sealed Sources (2003/122/Euratom), Directive on Outside Workers (90/641/Euratom), Directive on Public Information (89/618/Euratom). 2010.

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# Exposure of workers in France due to naturally occurring radioactive materials (NORM)

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## Abstract
According to the council Directive 96/29 Euratom, handling or storage NORM or TENORM has to be considered from the radiological protection point of view. This directive has been implemented in French regulations by, in particular, the Ministerial order of May 25, 2005 related to activities operating NORM not used because of their radioactive properties. It imposes radiological characterization of any materials and assessments of the effective dose received by workers to be done. Since the publication of this text, ASN and IRSN have already received ninety studies which provide information about activities of materials and occupational exposure of nine different types of industrial facilities. These data show that activity concentrations strongly vary according to the type of material and industrial activity. Waste generally contain the highest activity concentration of natural radionuclides. Activities in raw materials and products sometimes exceed the exemption level recommended by IAEA for the use of NORM. Activity of $^{226}$Ra and its daughters in waste or activity of $^{210}$Pb and its daughters in ashes and dust related to heating processes are sometimes greater than the activities of the other radionuclides of $^{238}$U series. Concerning occupational exposure, doses reported by operators range from less than 1 µSv.yr$^{-1}$ to 82 mSv.yr$^{-1}$. Based on the French feedback, the following conclusions can be drawn. Assessments are still expected for some industries. About 10% of calculated doses are greater than the effective dose limit of 1 mSv.yr$^{-1}$ for the public and need further examination. The highest doses correspond to the production of compounds with thorium. External and internal exposures are often of the same order of magnitude. Some types of industrial facilities currently not concerned by the French ministerial order, e.g. paper mills, are concerned by NORM and TENORM issue.

## Introduction
Some industrial activities such as ceramic production or coal combustion in thermal power stations involve the use of materials, usually regarded as non-radioactive, but containing naturally occurring radionuclides (NORM). Handling or storage of these materials can lead to a significant increase of occupational exposure. According to the

council Directive 96/29 Euratom (Council Directive, 1996), this matter has to be considered from the radiation protection point of view. French authorities implemented this directive to the French health and labour codes detailed in the Ministerial order of May 25, 2005 (Ministerial order, 2005). It sets out a list of ten types of industrial activities concerned by NORM and operators have to assess the effective doses received by workers on the bases of a radiological characterization of raw materials, by-products and waste involved in or produced by the technological processes. ASN asked IRSN to evaluate the methods adopted by some operators for assessing these doses and to draw the first conclusions in terms of radiological protection. Based on data presented by operators in the ninety studies received so far, a summary of the results of these studies in terms of activity concentrations and occupational exposure are presented hereafter.

## Material and methods

Since the publication of the Ministerial order of May 25, 2005 (Ministerial order, 2005), ASN and IRSN have already received ninety studies which provide activities of materials and doses calculated for nine types of industrial activities (cf. table I). The majority of the studies deals with occupational exposure and about 91% of them report on effective doses received by workers.

**Table 1. Breakdown of studies by different types of industrial activities.**

| Industrial activities sets by the French ministerial order of May 25, 2005 | Part (%) |
|---|---|
| Production of refractory ceramics and smelting, metallurgy and glass industry using them | 49% |
| Coal combustion in power plants | 16% |
| Production of zircon and baddeleyite, and smelting or metallurgy plants using it | 15% |
| Treatment of tin, aluminium, copper, titanium, niobium, bismuth and thorium ores | 7% |
| Production of phosphated fertilizers and phosphoric acid | 6% |
| Production or use of compounds with thorium | 3% |
| Treatment of lanthanides series and production of pigments containing them | 2% |
| Spas | 1% |
| Underground water treatment by filtration | 1% |
| Treatment of titanium dioxide | 0% |

Among the ninety studies, 43 present activity concentration measurements mainly carried out by gamma spectrometry for about 500 different samples. It represents more than 4200 results of measurements. Radionuclides of $^{238}U$, $^{232}Th$ and $^{235}U$ series and $^{40}K$ represent respectively 47%, 29%, 15% and 9% of measurements and 69% of the measurements for $^{238}U$ series or $^{232}Th$ series are above the detection limit (LD), 32% for $^{235}U$ series and 59% for $^{40}K$.

## Uranium and thorium series activity concentrations

### Measurements taken into account

Activity concentrations mainly measured by gamma spectrometry were analysed by IRSN. Uranium and thorium series have been divided into groups of radionuclides as recommended by the European Commission (Chen *et al*, 2003). For each chain segment, an activity was determined by the method whose principle is described hereafter (Maigret *et al*, 2008). For a given sample, if only one radionuclide activity of a group is measured, its activity stands for the group. If several radionuclide activities are measured and their measurements are consistent, the average of more reliable activities stands for the group. For example, regarding the chain segment of $^{226}Ra^{+1}$, deconvolution of the $^{226}Ra$ gamma ray (186.2 keV) and the $^{235}U$ gamma ray (185.7 keV) cannot be done accurately by the classic HP-Ge detectors (detectors made of High Purity Germanium) mainly used in studies, whereas the activity of $^{214}Pb$ or $^{214}Bi$ is easy to measure with those detectors. So the average of activities of $^{214}Pb$ and $^{214}Bi$ stands for the group of $^{226}Ra^{+}$.

### Activity levels and radioactive disequilibrium in NORM used in French industries

As a first step, all samples presented by operators were classified in four categories: raw materials, products, waste and environment. The category "Environment" represents a fifth of the samples and includes all the samples collected in the environment, e.g. soil, sediments, vegetables… These data have been excluded from our work. The category "Products" includes final products and by-products and the category "Waste" includes solid waste, effluents, sludge and dust. 26% of materials considered by operators are raw materials, 34% are products and 39% are waste.

In the second step, twelve industrial activities were identified: coal combustion, glass industry, production and use of zircon and baddeleyite, production of phosphated fertilizers, production of refractory ceramics, production or use of compounds with thorium, spas, treatment of aluminium ores, treatment of kaolin ores, treatment of lanthanides series, treatment of titanium ores and underground water treatment by filtration. All the samples were classified according to these categories.

Eventually, for each category of materials and for each chain segment, the minimum, the maximum and the median values, as well as the first and the third quartile values have been determined. Figure 1 presents activities of the most often measured chain segments: $^{226}Ra^{+}$ and $^{228}Ra^{+}$ groups.

[1] Symbol '+' after a nuclide denotes a segment chain headed by that nuclide, e.g. $^{226}Ra^{+}$ corresponds to this segment chain: $^{226}Ra$, $^{222}Rn$, $^{218}Po$, $^{218}At$, $^{214}Pb$, $^{214}Bi$ and $^{214}Po$.

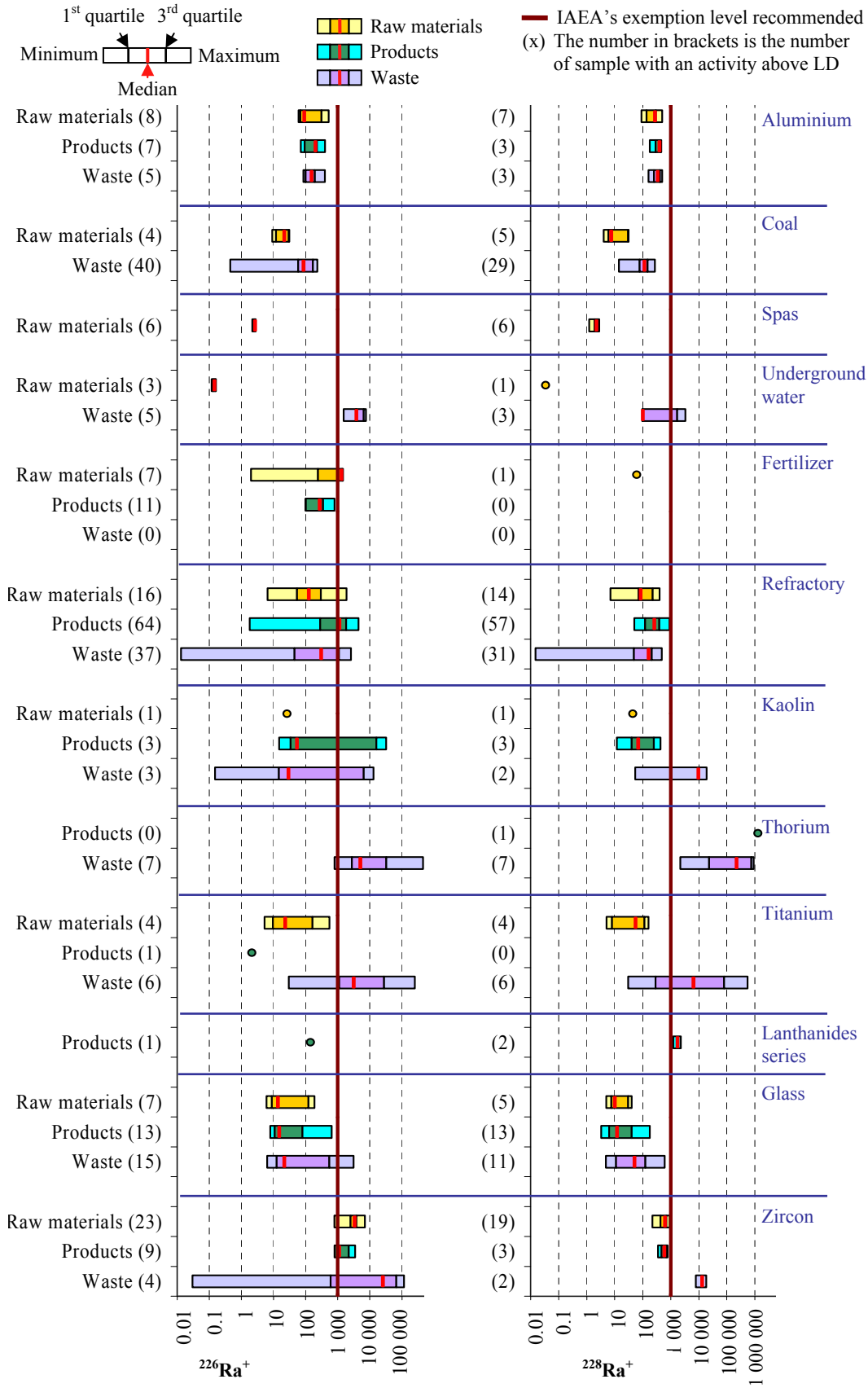


**Fig. 1. Activities of $^{226}Ra^{+}$ and $^{228}Ra^{+}$ ($Bq.kg^{-1}$).**

## Conclusions relative to activity levels

Based on French feedback, the following conclusions can be drawn:

- activity concentrations strongly vary according to the type of material and industrial activity;
- waste generally contain the highest activity concentration of natural radionuclides;
- 55% of materials with an activity higher than 1000 Bq.kg$^{-1}$, the IAEA's exemption level recommended for the use of NORM, are waste, 29% are products and only 16% are raw materials.
- the number of radionuclides measured and the accuracy of their activity determination are barely enough to assess the secular equilibrium or radioactive disequilibrium that occurs in NORM.
- imbalances have been identified: activity of $^{226}Ra^{+}$ group in waste (e.g. the category "underground water") or activity of $^{210}Pb^{+}$ group in ashes and dust related to heating processes is greater than the activities of the other groups of $^{238}U$ series. For example, figures 2 and 3 show two illustrations of excess of $^{210}Pb$ in dust from the refractory industries.

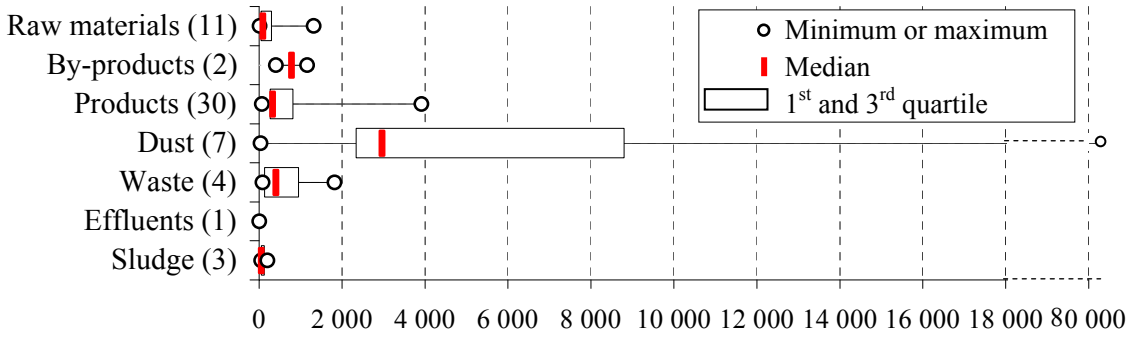


**Fig. 2. Activities of $^{210}Pb^{+}$ in the refractory industries (Bq.kg$^{-1}$).**

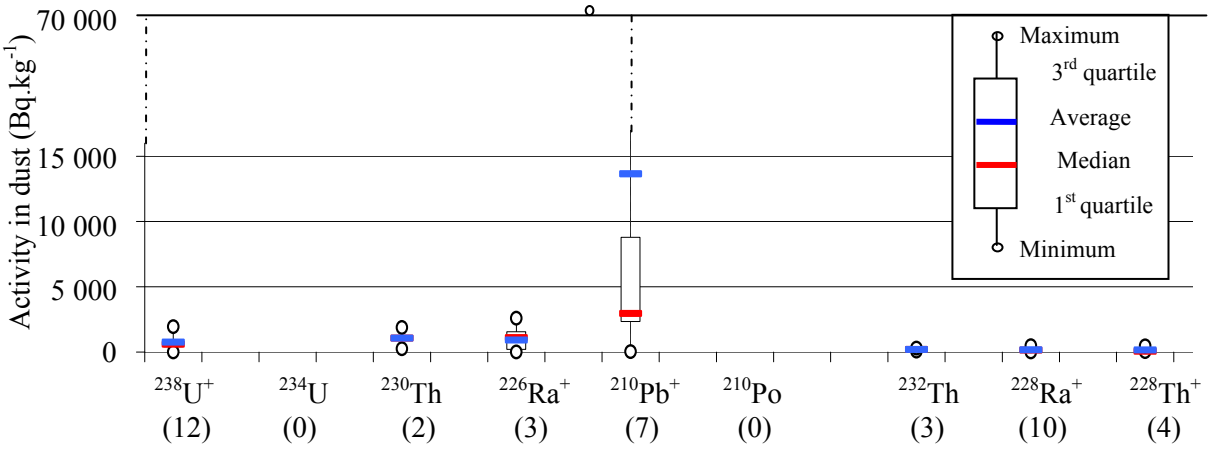


**Fig. 3. Activities of $^{210}Pb^{+}$ in the refractory industries (Bq.kg$^{-1}$).**

## Occupational exposures

### Data collected

The Ministerial order of May 25, 2005 imposes on operators to assess the occupational effective doses and to take into account external exposure, as well as internal exposure by dust inhalation and inhalation of radon progeny. Since the publication of this text, operators have assessed occupational exposure for more than 400 workplaces. A third of doses are less than 0.1 mSv.yr$^{-1}$, a half are less than 0.25 mSv.yr$^{-1}$ and 15% of doses are still greater than the effective dose limit of 1 mSv per year for the public in France (French Public Health Code, 2006) and so need further examination.

Firstly, some doses calculated by operators do not take into account external exposure or internal exposure by dust inhalation even if this route could be a significant pathway of exposure. Secondly, only some doses also consider exposure due to inhalation of radon progeny. Thirdly, only some operators take into account the exposure due to natural background in their assessment. Finally, the doses received by workers vary strongly according to the type of industrial facilities. Due to the different approach retained by operators, it is not possible to compare directly the doses reported. So, on the one hand, when data are available, exposure due to natural background was subtracted from the effective doses reported by operators. On the other hand, in order to compare the effective dose for each type of industrial activity and to identify the significant route of exposure, IRSN has analyzed the doses calculated for workers due to external and internal exposure for each type of industrial activity. Data relative to exposure to radon have been considered specifically.

### Effective doses above the natural background

#### *Production of refractory ceramics and smelting, metallurgy and glass industry using them*

The production of refractory ceramics and smelting, metallurgy and glass industry using them is the most often assessed industrial activity: more than 100 workplaces have been assessed. Figure 4 shows the added effective dose. For two workplaces, doses are greater than 1 mSv.yr$^{-1}$ with a maximum of 1.5 mSv.yr$^{-1}$ but these doses are still consistent with literature (NORM V, 2008).

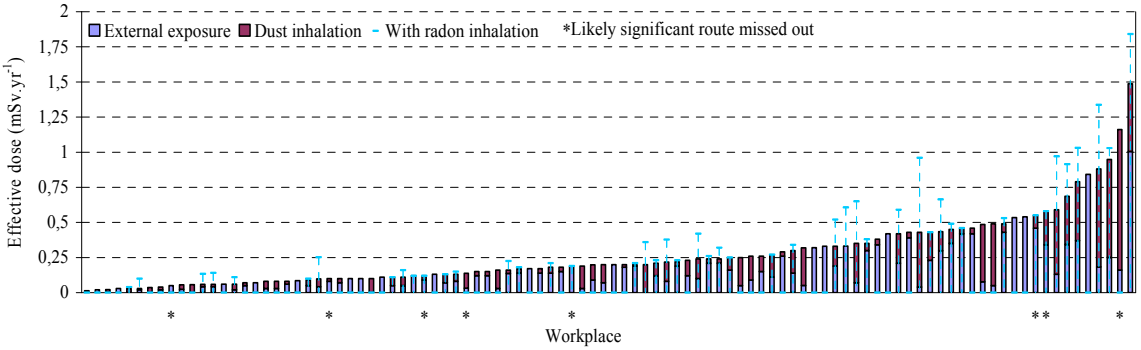


**Fig. 4. Production of refractory ceramics - Added effective doses.**

### *Production of zircon and baddeleyite, and smelting or metallurgy plants using it*

The production of zircon and baddeleyite, and smelting or metallurgy plants using it is the second most often assessed industrial activity: more than 60 workplaces have been assessed. Figure 5 shows the added effective dose reported by operators. For eight workplaces, dose is greater than 1 $mSv.yr^{-1}$ with a maximum dose of 2.3 $mSv.yr^{-1}$. This dose is still consistent with results published in literature for zircone production by a thermic process (NORM V, 2008). Moreover, it is worth mentioning that the highest doses correspond to two studies in which a hypothetic and maximum time of exposure of 1600 $h.yr^{-1}$ have been considered.

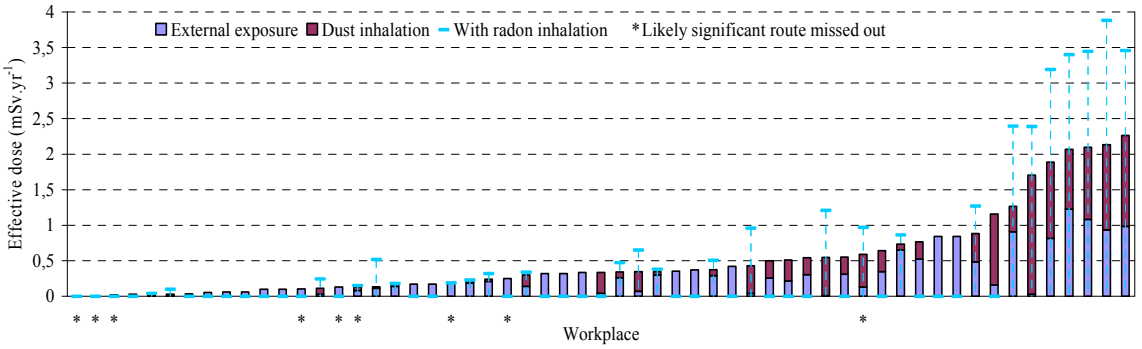


**Fig. 5. Production of zircon and smelting or metallurgy plants using it - Added effective doses.**

### *Treatment of tin, aluminium, titanium and niobium ores*

The treatment of tin, aluminium, titanium and niobium ores is the third most often assessed industrial activity: more than 40 workplaces have been assessed. Figure 6 shows the added effective dose. For 13 workplaces, doses are greater than 1 $mSv.yr^{-1}$ with a maximum of 6 $mSv.yr^{-1}$ and are consistent with literature (NORM V, 2008), (UNSCEAR, 2000).

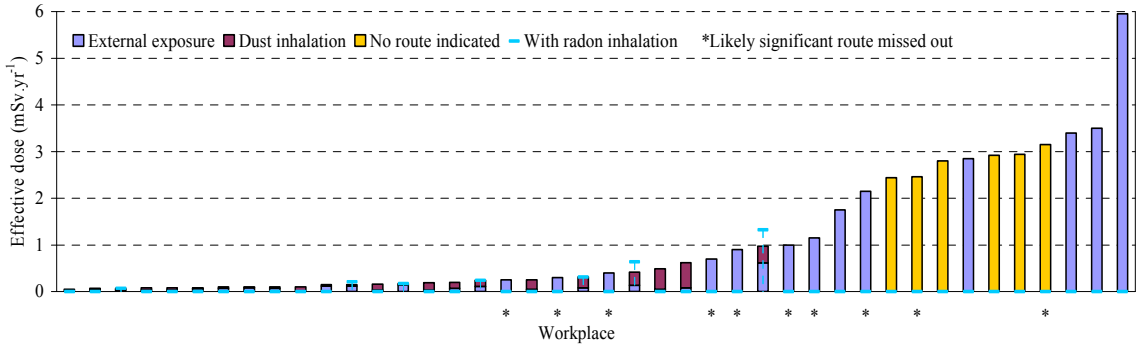


**Fig. 6. Treatment of ores (Sn, Al, Ti, Nb) - Added effective doses.**

It is worth mentionning that the highest doses correspond to only one study. New measurements in 2008 show an important reduction of dose rate: based on these new measurements, the maximum dose decreases to 3.2 $mSv.yr^{-1}$.

### *Coal combustion in power plants*

30 workplaces for coal combustion in thermal power plants have been assessed. Figure 7 shows the added effective dose reported by operators. For any workplace, dose is less than 1 mSv.yr$^{-1}$ with a maximum dose of 0.4 mSv.yr$^{-1}$ which is consistent with literature (Smith *et al*, 2001).

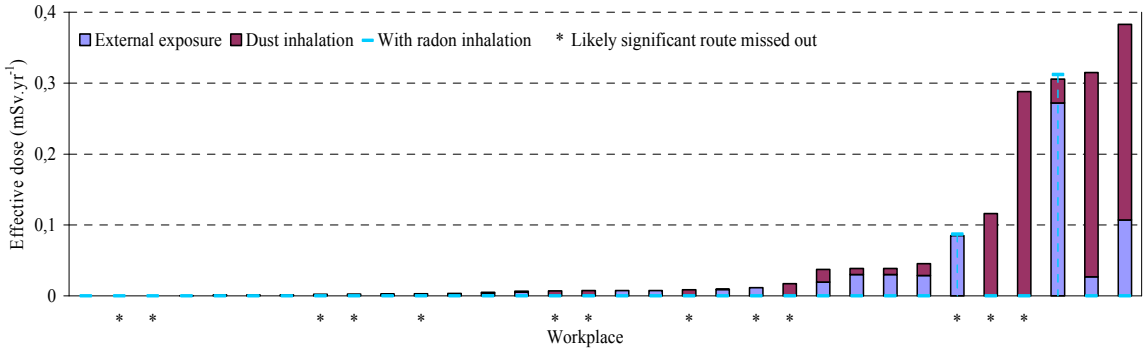


**Fig. 7. Coal combustion - Added effective doses.**

### *Other industrial activities*

For the production or use of compounds with thorium, six workplaces have been assessed. For two workplaces, the doses are greater than 1 mSv.yr$^{-1}$ with a maximum dose of 82 mSv.yr$^{-1}$. This dose is mainly due to dust inhalation. In order to reduce occupational exposure, the operator intended to equip workers with personal protective equipment and he should periodically clean the working planes and install a system of air filtration in his installations. These actions should significantly reduce the doses but IRSN and French authorities have not yet received the new study.

For the production of phosphated fertilizers, six workplaces have been assessed. Every doses are less than 1 mSv.yr$^{-1}$ with a maximum dose of 0.5 mSv.yr$^{-1}$ which is consistent with literature (NORM V, 2008), (AIEA, 2006).

For the treatment of lanthanides series, three workplaces have been assessed. Every doses are less than 1 mSv.yr$^{-1}$ with a maximum dose of 0.3 mSv.yr$^{-1}$ which is consistent with literature (AIEA, 2006).

### Route of exposure

For only four types of industrial activities, workplaces have been sufficiently studied to allow to identify the significant route of exposure. Figure 8 shows the contributions of external and internal exposure for these four types of industrial activities.

– For coal combustion, data show clearly that external exposure is the significant route of exposure.
– For the production of refractory ceramics and smelting, metallurgy and glass industry using them and the production of zircon and baddeleyite, and smelting or metallurgy plants using it, data collected do not clearly show any significant route of exposure, though external exposure seems to be the most important pathway.
– For treatment of ores, no significant route of exposure could be identified.

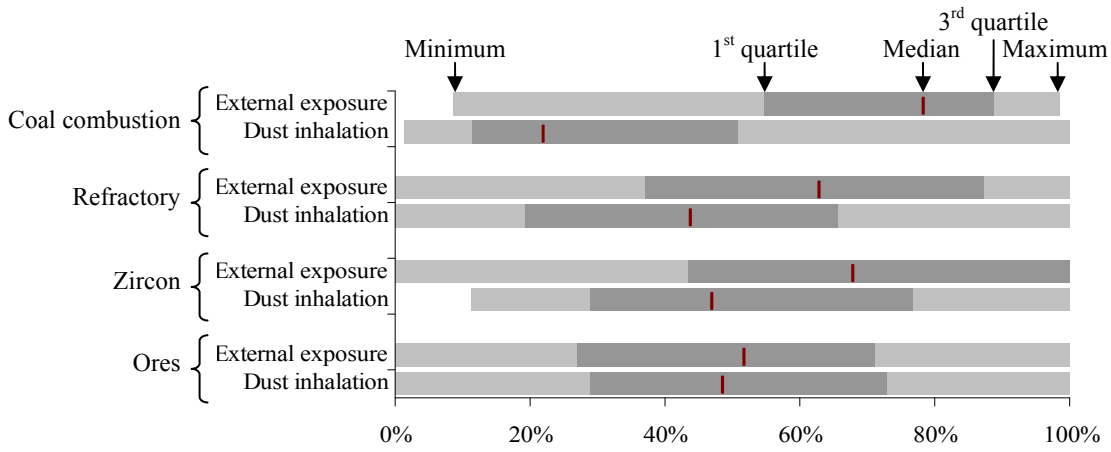


**Fig. 8. Contributions of external exposure and dust inhalation.**

## Conclusions

Based on the feedback gained by IRSN and French authorities from the studies conducted by operators, the following conclusions can be drawn additionally to those presented in (Maigret *et al*, 2008):

- assessments are still expected for some industrial activities referred to in the Ministerial order of May 25, 2005 (Ministerial order, 2005). For example, in the next future, studies dealing with occupational exposure due to underground water treatment by filtration should be carried out;
- about 10% of added effective doses are greater than 1 mSv per year and need further examination;
- the highest doses were found in facilities which produce materials involving thorium (82 $mSv.yr^{-1}$) and which treat tin, aluminium, titanium and niobium ores (6 $mSv.yr^{-1}$). For the other types of industrial activities, the maximum dose remained below a few mSv per year;
- external and internal exposure are often of the same order of magnitude;
- some types of industrial facilities currently not referred to in the French ministerial order, e.g. paper mills, are concerned by NORM and TENORM issue and could be added to the list of industrial facilities set by the Ministerial order of May 25, 2005 (Ministerial order, 2005).

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# Radiological baseline study on the planned Sokli phosphate mine in Finnish Lapland

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## Abstract
Results of the radiological baseline study on the planned Sokli phosphate mine in Northern Finland are presented. From radiation protection point of view, the Sokli carbonatite massif contains considerable amounts of natural radioactive substances, i.e. thorium and uranium and their decay products. These substances are especially rich in the niobium ore. In connection with the Environmental Impact Assessment, the Radiation and Nuclear Safety Authority (STUK) carried out the radiological baseline study at the site of the planned Sokli phosphate mine and its surrounding environment. In the baseline study the activity concentrations were studied in ecosystems at the Sokli site, as well as in the immediate vicinity where the mine may have some radiological impact. The objective of the study was to obtain a detailed understanding of the radiation levels at and in the vicinity of the Sokli mining site before the commencement of mining and treatment of phosphate ore. The baseline study can be used in the future to assess possible impacts on the environment of the natural radionuclides released during the mining and milling process.

## Introduction
The Sokli carbonatite massif is part the Kola alkaline province which covers an area of approximately 100,000 km$^2$, and is one of a few regions in the world where the paleozoic ultramafic-alkaline magmatism is well developed. A characteristic feature of the Kola carbonatites in many complexes is their close relation with phoscorites, rocks consisting essentially of apatite, magnetite and forsterite or diopside (Mi Jung Lee, et al. 2006).

Sokli is located in Savukoski municipality in the Finnish Lapland, close to the border with Russia (Fig. 1). The mineral deposit is associated with a carbonatite massif some 360 million years old, with a land surface diameter of 5–6 km. In addition to the phosphorus minerals, the carbonatite massif contains other ores, such as iron, niobium and vermiculite minerals. The Sokli carbonatite massif has been explored since 1967 and the pilot mining and milling of phosphorus minerals was performed in late 1970's. However, in those days the commencement of phosphate ore concentrate and phosphate

products in an industrial scale was not profitable. Today the plans for the Sokli mine project include opencast ore extraction and treatment, crushing and grinding of the ore, beneficiation, dewatering and storage of the products, tailings storage, water treatment and auxiliary activities. The planned production is 2.0 million tonnes of phosphorous concentrate per year. The amount of ore to be mined will be 4-10 million tonnes per year depending on the mining and milling strategy in the area. There are also plans to extract iron and other valuable minerals later. According the planned capacity, it has been estimated that the richest phosphorus ores will last for around 20 years of production. If the poorer parts of the deposit are exploited, it may be possible to extend the life time of the mine by twenty to thirty years, or even more.

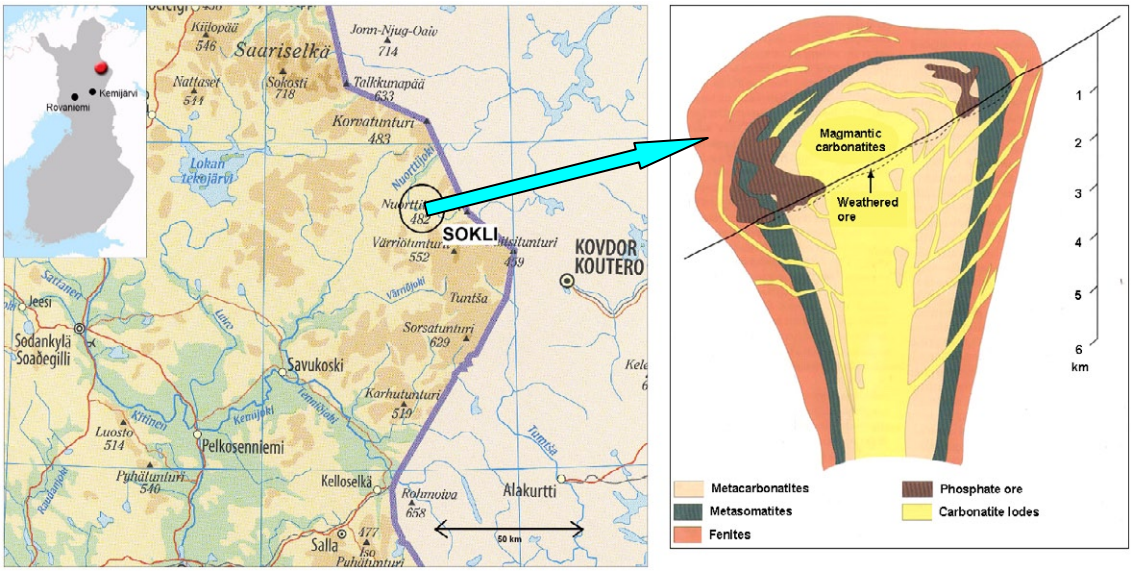


**Fig. 1. Location and shape of the Sokli carbonatite massif. The magmatic carbonative ore continues tens of kilometers below the ground level.**

## Material and methods

The environmental samples were collected in 2008 and 2009. The samples were pre-treated and analysed at the STUK's Regional Laboratory in Northern Finland. In-situ gamma spectrometric measurements and radiochemical analyses of uranium in water samples were made by STUK's laboratories in Helsinki. Different environmental samples were collected: river water, river sediment, fishes, lichen, beard moss, water moss (*Fontinalis antipyretica*), mushrooms, berries, reindeer meat, ground water, soil, moose meat and pochard.

The environmental samples were analysed for gamma-emitting radionuclides with HPGe gamma spectrometers. The concentrations of $^{238}$U, $^{235}$U, $^{226}$Ra, $^{228}$Ra, $^{228}$Th, $^{232}$Th, $^{210}$Pb and $^{40}$K were analysed by using containers, which were inside the gas-tight aluminium bags in vacuum. The samples were kept in these bags at least three weeks before measurements so radium, radon and its short lived decay products were in equilibrium when measured. The ground water samples were also studied for $^{222}$Rn.

In addition, radiochemical analyses were done to analyse activity concentrations for $^{210}$Po and $^{210}$Pb. All the solid samples were first dried at 105ºC and then

homogenized. Suitable sample quantities were spiked with $^{209}$Po tracer and digested by microwave. Polonium was deposited on silver plate and measured by alpha spectrometer (Vesterbacka and Ikäheimonen 2005). All samples were analysed before 6 months has gone from the sampling date. The solution remaining from the first deposition was stored about 6 months to allow the in-growth of $^{210}$Po from $^{210}$Pb. The second $^{210}$Po deposition was then carried out by using $^{208}$Po tracer and the $^{210}$Po activity was measured by alpha spectrometer. The $^{210}$Pb content of the samples was calculated and the first $^{210}$Po determination was corrected for the radioactive decay between the time of sample collection and analysis and the in-growth of $^{210}$Pb during sample storing before first $^{210}$Po deposition was subtracted.

## Results

The highest radioactivity concentrations were measured in the niobium ore and in the old mill tailings remaining at the site from the pilot enrichment in late 1970's. In the niobium ore the radioactivity concentrations were 8500 Bq/kg for $^{232}$Th and 2200 Bq/kg for $^{238}$U. In river sediments the activity concentrations of these radionuclides varied from few Bq/kg up to three hundred Bq/kg. In reindeer meat, game meat, berries, mushrooms and in river waters the activity concentrations remained in nearly all the samples below the detection limits of the used gamma spectrometric methods. More precise radiochemical analyses of uranium isotopes were performed for different water samples. The lowest concentrations were found in river water (0.001 - 0.024 Bq/L) and the highest concentration in spring water (0.01 - 0.02 Bq/L). The results are shown in Table 1.

$^{210}$Pb and $^{210}$Po were found in all analysed samples. The lowest values were found in river waters and the highest values in water moss (*Fontinalis antipyretica*) which is a common aquatic plant in Lapland and known to be a good bio-indicator for heavy metals. High amounts of $^{210}$Pb and $^{210}$Po were found also in reindeer lichen (*Cladina rangiferina*) which is the main food plant of reindeers and in beard moss (*Bryoria*) growing on spruce and pine branches and in bolete. $^{210}$Pb and $^{210}$Po concentrations in different environmental media are presented in Table 2.

The activity concentrations of $^{222}$Rn in ground water samples varied between 30-110 Bq/L. In river water samples radon concentrations were below detection limit 30 Bq/L.

**Table 1. Concentrations of $^{234}$U and $^{238}$U in different water samples collected at the planned phosphate mine and its surroundings.**

| Water type | U-234, Bq/L | | U-238, Bq/L | |
|---|---|---|---|---|
| | Year 2008 | Year 2009 | Year 2008 | Year 2009 |
| River water | 0.0044 - 0.0241 | 0.0011 - 0.0099 | 0.0033 - 0.0199 | 0.0017 - 0.0088 |
| Spring water | 0.0189 | 0.0116 | 0.0203 | 0.0128 |
| Ground water | 0.0014 ja 0,0054 | 0.0011 ja 0.0045 | 0.0028 ja 0.0081 | 0.002 ja 0.0059 |

**Table 2. Concentrations of $^{210}$Po and $^{210}$Pb in different environmental samples collected at the planned phosphate mine and its surroundings.**

| Sample | $^{210}$Po, Bq/kg | $^{210}$Pb, Bq/kg |
|---|---|---|
| Ground water | 0.03 - 0.04 | 0.008 - 0.03 |
| River water | 0.003 - 0.01 | 0.0004 - 0.01 |
| Bolete | 129 - 832 | 1.9 - 3.4 |
| Milk caps | 15.6 - 42.9 | 9.9 - 21.7 |
| Russule | 6.4 - 18.3 | 4.1 - 6.7 |
| Trout, grayling | 2.0 - 19 | 1-3.6 |
| Cloudberry | 1 - 1.4 | 1.4 - 1.6 |
| Lingonberry | 4.6 - 4.9 | 2.5 |
| Blueberry | 4 | 2 |
| River sediment | 15 - 294 | 7 - 118 |
| Water moss | 182 - 1190 | 47 - 659 |
| Reindeer lichen | 167 - 186 | 89 - 96 |
| Beard moss | 302 - 472 | 166 - 251 |
| Moose meat | 4.3 - 4.7 | 0.7 - 1 |
| Reindeer meat | 8.0 - 83 | 0.8 - 2.5 |

## Conclusions

The radiological baseline study in the area of the planned phosphate mine and concentration plant was performed in 2008-2009. It was considered important to investigate the natural radiological situation of the site and its environment before the commencement of mining and treatment of the phosphate ores to be able to assess possible radiological impact of these activities in the future. The Sokli carbonatite massif contains various ores and some of them contain elevated amounts of thorium and uranium. Radiological studies done before any industrial actions are the only way to verify the results of the Environmental Impact Assessment (EIA) because the studied radionuclides are occurring naturally in the environment. The local people have been worried about the radiological impact of the mining and enrichment activities to their health and the environment. It is important also to them to know the natural levels of radioactivity in different environmental media.

Preliminary results of the baseline study show that the amounts of natural radionuclides in mushrooms, berries, fish, lichens, beard moss, reindeer meat and moose meat samples were at the same level as in other environmental samples analysed from Finnish Lapland. The activity concentrations of $^{234}$U, $^{238}$U, $^{210}$Pb and $^{210}$Po in river water were close to the average concentration of Finnish tap water from waterworks 0.02. 0.015. 0.003 and 0.003 Bq/L, respectively. The highest $^{210}$Pb and $^{210}$Po concentrations were found in bolete, water moss, beard moss and lichen, which are good bio-indicators for radionuclides.

Radioecological investigations were performed at the Sokli area also in late 1970's when earlier plans to start phosphate production was current. The results obtained in 1970's were very similar compared with the results of this study. This indicates that the pilot production activities in 1970's had no significant radiological impact to the environment.

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# Naturally occurring radionuclides in drinking water – An overview of the problem in Sweden

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## Abstract

In Sweden, uranium rich soils and bedrocks can give rise to high concentrations of naturally occurring radionuclide (NORM) in groundwater. If the extracted water is then used as domestic water supply, an appreciable radiation dose can be received. Radionuclides that are of interest, originate principally from the decay series of uranium ($^{238}$U) and include radon ($^{222}$Rn), radium ($^{226}$Ra), polonium ($^{210}$Po), lead ($^{210}$Pb) and $^{238}$U itself if the latter is present in high concentration (> 100 µg/l). A recent country wide mapping of about 700 drilled wells reveal that geological criterion cannot alone explain concentration variations of radionuclides in groundwater. 8% of the studied wells, located outside regions containing uranium rich bedrocks, had a radon concentration exceeding the recommended action level of 1000 Bq/l in water. A proper understanding of the factors guiding the mobilisation and transport of decay products of $^{238}$U in the subsurface is a prerequisite to make accurate risk prediction. There are about 250 000 drilled wells that are used permanently in Sweden and between 5000 –10000 wells get constructed every year. The concentrations of radionuclides except for radon are seldom analysed. The National Board of Health and Welfare in Sweden recommends radon measurements in drinking water coming from private wells. Results of water analyses, if or when performed are not communicated to a centralised database that can be used by various national authorities but efforts are ongoing to build such a system. Knowledge regarding the extent to which private well owners check the quality of their water with regards to radionuclide and take appropriate remediation measures is poor. An ongoing survey regarding remediation measures is hoped to provide additional information on the issue.

# NORM in the petroleum and geothermal industries: evolution of the French radioprotection legislation

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## Abstract

Fluids produced from oil or gasfields and aquifers are naturally radioactive due to the presence of potassium-40 and isotopes from decay chains of uranium-238 and thorium-232 in these reservoirs. Thus, natural radionuclides can be carried away towards surface when petroleum or geothermal reservoirs are drilled and produced. These radionuclides can precipitate in deposits forming in process equipment, which can then be unusually radioactive, expose workers to hazardous materials and create waste disposal problems. Workers can also be exposed to high concentrations of radon. Due to their natural origin, these accumulations of radionuclides in equipment are called Naturally Occurring Radioactive Materials (NORM). They are also known as TENORM, because they are technologically enhanced.

In closed systems, exposition to gamma radiations penetrating equipment to the external surface can be reduced by restricting and controlling access to appropriate areas. But personnel may come into direct contact with NORM during maintenance and cleaning operations, when opening equipment and vessels.

In France, hygiene and safety rules concerning the protection of workers in the petroleum and geothermal industries are formulated in a specific legislation related to extraction and mining. Presently, both petroleum and geothermal industries are not included in the list of activities which have to follow radioprotection measures given by this legislation. French authorities are currently planning to update the list by including these two activities. There is a real need to realize a state-of-the art of the exposition level of French workers and to match the dose limits to those given by the Labor and Public Health Codes.

This paper will present the origin of NORM and radon contamination in the petroleum and geothermal industries and discuss the French legislation evolution for the radioprotection of personnel working in these two industries.

# Ukrainian experience of monitoring of radiation exposure of population determined by building materials

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## Abstract
This article contains results of monitoring of the specific radioactivity of the natural radionuclides in the minerals used as construction raw materials. In total, 22 quarries (opencast mines) of crushed stone (gravel), 10 quarries of sand and 4 of clay were studied and around 800 samples of mineral raw materials used in construction were analyzed. Effective doses of external radiation exposure of the population determined by building raw materials were defined.

It was found that the raw materials produced at 12% of the quarries do not comply with the relevant requirements of the national regulations, thus, such materials can not be used for the residential construction. Based on the assessment of the study, it was found that the average weighted effective exposure dose of the population resulting from radioactivity of the construction materials in the structure of the overall country housing is 0.23 mSv per year.

## Introduction
External exposure of population to radiation in the houses is determined by naturally occurring radioactive materials contained in building materials. Studies of this source of radiation are currently done in various countries of the world, however, there is less data for radiation exposure levels inside houses than that for the outside, at the open space. This is related to the fact that there are no relevant regulations restricting exposure of population in the houses to radiation in the majority of the countries of the world. The most profound review of the results of research of such a source of radiation is quoted in the report of the United Nations Scientific Committee on the Effects of Atomic Radiation [1]. Thus, based on the Committee data, the average weighted radiation exposure dose of the population of the planet is in the range between 20 nGy to 200 nGy per hour, with the average level of 84 nGy per hour.

Furthermore, according to the Report [1], the lowest levels of radiation exposure were registered in New Zealand, Iceland and the United States, where it was lower the 40 nGy per hour level, whereas the highest (95 to 115 nGy per hour) were registered in

Hungary, China, Portugal, Austria, Spain and Iran. As a rule, the high radiation population exposure levels exist due to the use of stone and concrete with higher content of naturally occurring radionuclides in construction of housing. [2,3]. For example in Sweden lots of dwellings were constructed of light concrete before 1975. The raw material used for the production of light concrete contains large volumes of alum shale, rich in Uranium. The average dose to the dwellers of the light concrete houses is 1,1 mSv/a.

First regulations restricting the content of the naturally occurring radioactive materials (NORM) in building materials in the USSR have had come into being in 1976 [4]. This regulatory standard was developed by the specialists of the Leningrad Scientific and Research Institute for Radiation Hygiene then headed by Ye. Krisyuk. His monograph manuscript provided the scientific rationale for the methodological specifics related to substantiation of the numerical values of Radium-226, Thorioum-332 and Potassium-40 content in construction materials. These standards [norms] remain the same to date and are applied according to Norms of Radiation Safety of Ukraine, thus, it is worthwhile to look at those in greater detail.

Here is the main idea behind such standard. The value of the standard [norm] is no more than 370 Bq/kg of the specific effective radioactivity ($C_{eff}$) of the naturally occurring radionuclides (Radium-226, Thorioum-332 and Potassium-40) contained in construction materials. This was 4 times more the relevant average value at the territory of the USSR (93 Bq/kg). Expected excess of the standard throughout the whole territory of the country was around 1%. Effective external radiation exposure dose in the houses built with the use of construction materials with NORM value of $C_{eff} = 370$ Bq/kg is 1.75 mSv per year for the period of being inside a house of 7000 hours per year [6].

Construction materials with the higher content of NORM are permitted to be used in road construction (up to 740 Bq/kg within the limits of towns and other settlements and between 740 Bq/kg and 1850 Bq/kg beyoned such limits) [7].

Monitoring of the NORM content of construction materials and mineral raw materials used in construction is mandatory in Ukraine throughout the whole territory. The article includes the analysis of the study results for 95% of all quarries in Ukraine where construction materials and mineral raw materials used in construction are produced. Based on data, resulting from the direct studies, effective radiation exposure doses assessment was made for the population of different regions of the country.

## Material and methods

### Measurement method

Spectrometry method involving the use of ORTEK (US) high resolution gamma spectrometry system with NaI (T1) scintillation detector and well was applied in the framework of this article in order to do the measurements of mineral construction raw materials samples.

Measured samples were put inside Marinelli container, that fill the space of the well with the detector crystal completely. With this placement of samples the geometrical effectiveness of registration is substantially higher than when cylinder form crystals are used.

Large volume well, 150 mm diameter, 150 mm height, was used to provide for sufficient sensitivity. AMERSHAM (Germany) standard source was used for calibration of the gamma spectrometer.

For geometry 'Marinelli 1 litre' the value of the lowest detectable radioactivity was 2 Bq/kg for Thorium-232, 3 Bq/kg for Radium-226 and 10 Bq/kg for Potassium-40.

Effective specific radioactivity of a sample ($A_{eff}$) was calculated based on the values of specific radioactivity of the NORM, Thorium-232, Radium-226 and Potassium-40.

$$A_{eff} = A_{Ra} + 1{,}31 \cdot A_{Th} + 0{,}085 \cdot A_K,$$

Where $A_{Ra}$, $A_{Th}$, $A_K$ – values of specific radioactivity of Radium-226, Thorium-232 and Potassium-40, Bq/kg.

### Method dose calculation

Transition coefficients from air-absorbed dose to effective dose and parameters for time of humans being enclosed inside house, quoted in attachments to the United Nations Scientific Committee on the Effects of Atomic Radiation report, were used to assess the average annual effective dose values [1]. Transition coefficient from air-absorbed dose to effective dose was set at the value equal to 0,7 Sv/Gy whereas factor for length of time of being inside housing was 0.8 [1].

The following data was used in relation to the country overall housing structure to do the assessment of the average weighted population effective radiation exposure doses (obtained from Radon measurement certificates from IHME (Institute of Hygiene and Medical Ecology) databases: buildings currently in operation (28 000 buildings) and building in process to be commissioned (34 000 premises). The certificate contains information about materials, used to construct a building, year of construction, number of residents in a building as well as that on the number of floors, coverings of the walls, type of dividers between floor and space under floor, availability of basements and ventilation system in buildings. Such information makes that possible to do adjustments when calculating values of the effective doses of radiation exposure of population considering specifics of architectural and layout solutions pertinent to individual regions in Ukraine.

For instance, housing structure of the rural areas of the Kiev Oblast comprises 60% of the building made of bricks, 33% are wooden buildings, 2% - adobe brick buildings and around 1% was build with the use of shell limestone and slag stone.

Effective exposure doses were calculated separately for residents of the brick-made and concrete-plate buildings in order to assess average annual radiation exposure doses of the urban population, and then the resultant value was weighted against the housing structure.

## Results

Results of measurements of the NORM content in samples of mineral construction raw materials from 22 crushed stone, 10 sand and 4 clay quarries were analyzed to assess relevant effective exposure doses.

It was found in the analysis that the average weighted specific radioactivity $\bar{A}_{eff}$ for selected quarries (580 samples) was 238 Bq/kg with standard deviation of 128 Bq/kg.

Relevant value for 10 sand quarries (80 samples) was 127 Bq/kg with the standard deviation 309 Bq/kg, and for 4 clay quarries (120 samples) it was 79 Bq/kg and 14 Bq/kg accordingly. Frequency distribution of the effective specific radioactivity values of the NORM was of the lognormal character.

Average value $A_{eff}$ for granite quarries usually does not exceed 370 Bq/kg and corresponds to Class I for mineral construction raw materials set out in NRBU-97 (Norms of Radiation Safety of Ukraine) (para 8.5.1.). Findings of the study have shown that $A_{eff}$ for the two crushed stone quarries (9% of the total number of all measured samples) exceeds that value, in other words, the mineral materials from such quarries can not be used for the residential construction. At present, such mineral materials are used for industrial and road construction.

According to the study, mineral raw materials produced at only 12% of all quarries of the country can not be used for the purposes of residential construction.

Tables 1-3 indicate data on NORM content in the mineral construction raw materials produced in separate regions of Ukraine.

**Table 1. Average Values of NORM Specific Radioactivity Samples of Crushed Stone from Quarries in Ukraine.**

| Oblast (Region) | Specific Activity, Bq/kg | | |
|---|---|---|---|
| | Radium-226 | Thorium-232 | Potassium-40 |
| Vinnitsya | 32 | 120 | 917 |
| Dnipropetrovsk | 143 | 420 | 1032 |
| Zhytomir | 45 | 72 | 1250 |
| Kiev | 42 | 131 | 1351 |
| Kirovohrad | 55 | 120 | 1217 |
| Rivne | 44 | 45 | 840 |
| Sumy | 99 | 69 | 1246 |
| Khmelnitsky | 44 | 52 | 976 |

**Table 2. Average Values of NOR Specific Radioactivity Samples of Sand from Quarries in Ukraine.**

| Oblast (Region) | Specific Radioactivity, Bq/kg | | |
|---|---|---|---|
| | Radium-226 | Thorium-232 | Potassium-40 |
| Zhytomir | 16 | 34 | 270 |
| Kiev | 6 | 7 | 88 |
| Volyn | 22 | 13 | 157 |
| Rivne | 28 | 33 | 401 |
| Chernihiv | 13 | 24 | 495 |

**Table 3. Average Values of NORM Specific Radioactivity Samples of Clay from Quarries in Ukraine.**

| Oblast (Region) | Specific Radioactivity, Bq/kg | | |
|---|---|---|---|
| | Radium-226 | Thorium-232 | Potassium-40 |
| Dnipropetrovsk | 33 | 43 | 447 |
| Zhytomir | 13 | 27 | 255 |
| Kiev | 21 | 33 | 481 |

## Radiation exposure doses

External radiation exposure doses of Ukraine's population are calculated based on results of the NORM specific radioactivity measurements of mineral construction materials samples, depending on the housing structure of regions and type of construction material. Table 4 contains results of effective radiation exposure doses calculation for population of Kiev Oblast as an example.

**Table 4. External Radiation Exposure Doses of Population Determined by NORM Contained in Building Materials.**

| Raw Material | Raw material use Coeff. | Absorbed Dose for Weight Fractions for Raw Materials in Buiding Materials wm, nGy/hour | | | | ED with wm = 0,5, mSv/a |
|---|---|---|---|---|---|---|
| | | wm = 1 | wm = 0,75 | wm = 0,5 | wm = 0,25 | |
| Crushed stone | 2,09 | 168 | 126 | 84 | 42 | 0,41 |
| Clay | 0,67 | 54 | 40 | 27 | 13 | 0,13 |
| Sand | 0,14 | 12 | 9 | 6 | 3 | 0,03 |

Table 5 illustrates main results of the assessment of external radiation exposure doses to population determined by construction materials. It was found that average annual radiation exposure dose for the urban population of Ukraine (around 70% of the total population of the country) is in the range of 0,28 – 0,33 mSv/a and that such doses are practically no different for residents of both brick-made and concrete-plate-made buildings. Only the mineral construction materials of the Class 1 were used to construct such buildings.

**Table 5. Effective Radiation Exposure Doses of Population of Selected Oblasts of Ukraine Determined by Radioactivity of Construction Materials, mSv/a.**

| Oblast | Urban Population | Rural Population |
|---|---|---|
| Vinnitsya | 0,31 | 0,10 |
| Volyn | 0,29 | 0,09 |
| Dnipropetrovsk | 0,29 | 0,30 |
| Zhytomir | 0,27 | 0,30 |
| Zaporizhzhia | 0,27 | 0,11 |
| Kiev | 0,33 | 0,20 |
| Odesa | 0,31 | 0,12 |
| Poltava | 0,31 | 0,09 |
| Rivne | 0,22 | 0,07 |
| Kherson | 0,28 | 0,09 |
| Cherkassy | 0,31 | 0,21 |

Effective radiation exposure doses may vary by several times for rural population depending on a specific region. For example, most of the dwellers in Volyn and Rivne Oblast live in buildings made of wood, thus, effective dose for the residents of the above Oblasts is less than 1 mSv per year. In Dnipropetrovsk, Kiev or Zhytomir Oblast houses are, as a rule, made of bricks, therefore, effective radiation exposure dose for population there is 2-3 times higher.

Thus, average weighted effective external radiation exposure dose for the population of Ukraine in the context of the country housing structure is 0.23 mSv/a.

It is still worthwhile to note out that in the separate instances, and, in particular, with regard to residents of the old concrete-panel type buildings, constructed back in 1950-1960 (prior to implementation of restrictions on the use of construction materials) individual effective radiation exposure doses may be much greater. According to our assessment, in such instances the maximum external radiation exposure dose is 1-1.5 mSv per year.

Such doses may be even higher than that in some separate cases in the rural areas. This is related to the use of the high NORM content waste produced by the local industrial facilities as additional material utilized to underlay foundation of a building. So, individual privately owned houses in Dniprodzerzhinsk is an example, in construction of which process waste of uranium milling was used. Individual radiation exposure doses of residents of such houses may be several times higher than average throughout the whole country.

Some outstanding cases have no impact on the assessment of the average radiation exposure doses, however give rise to serious concerns on the part of the local public when such cases are identified.

In summary of the results it can be stated that the existing mandatory radiological monitoring of the NOR in mineral construction materials and raw materials has made impossible the use [of non-compliant materials].

## Conclusions

1. Effective specific radioactivity $\bar{A}_{eff}$ of crushed stone is 238 Bq/kg for 22 analyzed quarries. Relevant value for 10 sand quarries was 127 Bq/kg and for 4 clay quarries was 79 Bq/kg.
2. Construction raw materials produced at 12% of the studied quarries can not be used for residential construction as effective specific radioactivity of the naturally occurring radionuclides in the relevant samples exceeds 370 Bq/kg.
3. Average weighted effective radiation exposure dose determined by radioactivity of construction materials for Ukraine's population with regard to the country's overall housing structure is 0,23 mSv/year.
4. Contribution of that component in the average annual radiation exposure dose of the country's population is 2% approximately.
5. Restrictions with regard to content of natural radionuclides in the mineral construction raw materials and materials used in residential construction have practically excluded that component from the category of sources that impact the value of radiation exposure risks for the population of the country.

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# The International Association of Oil & Gas Producers (OGP) naturally occurring radioactive material management (NORM) guidelines

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## Abstract
It has been established that Naturally Occurring Radioactive Materials (NORM) may accumulate in the oil/gas production process. This can create a radiation hazard for workers, the general public and the environment if adequate controls are not established. Regulations and guidelines on radiation protection in general and NORM in particular have been issued by various National and International entities, however these are not specific to the Oil & Gas industry, and do not provide clear and concise guidance to enable NORM to be managed effectively. The International Association of Oil and Gas Producers (OGP) established a task force, headed by Saudi Aramco, to develop NORM Management Guidelines. This task was completed after research, review and thorough consideration of most available NORM regulations, and the industry's current best practices in NORM management. The OGP guidelines offer a simple and logical method of managing NORM impacted operations that are flexible enough to be implemented across the industry. This paper outlines major aspects of OGP NORM management guidelines and elaborates on key issues related to the management of NORM in the Oil & Gas Industry, in particular on NORM monitoring, control of NORM contaminated equipment, managing NORM waste handling and disposal, and worker protection, awareness and training.

# Technologically enhanced NORM and heavy metals in iron and steel industry

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## Abstract

Iron and steel industry was ranked as the largest industrial source of toxic environmental contamination in USA. About 2 – 4 tones of various solid wastes (slag, sludge, dusts and scales) are generated per ton of steel produced. These wastes contain a notable concentration of Naturally Occurring Radioactive Materials – NORM and heavy elements that could be a source of environmental contamination and occupational exposure. Composite samples of different iron and steel industry's wastes were collected from four iron and steel factories. Natural radionuclides ($^{238}$U, $^{232}$Th, $^{40}$K, $^{210}$Pb, $^{210}$Po) and trace elements (e.g. Cd, Cu, Pb and Zn) were measured using gamma-ray spectrometry, alpha particle spectrometry and ICP-MS analytical techniques. There is a wide range of variation in the concentration of natural radionuclides and other elements that depends on their concentration in the process input materials and thermal treatment process. Occupational dose due to dusts inhalation was calculated. According to the assumed scenario, the occupational exposure is much lower than the reference dose limit. The environmental impact due to wastes storage and/or usage should be considered generally and case by case.

# NORM and trace elements fractionation in phosphate rock beneficiation processes: potential hazards and useful applications

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## Abstract

Beneficiation processes (mainly crushing, washing, magnetic separation and wet screening) of phosphate ore rock aims at increasing the phosphate content percentage. It starts with the ore rock and end with wet rock, and different rejects or processes by-products. By-products have potential hazards due to their content of Naturally Occurring Radioactive Materials – NORM especially uranium-238 series and heavy metals, e.g, Cd, Cu, Pb and Zn. They have also potential industrial and agricultural useful applications due its physical and chemical properties such as their relative high content of clay and iron. Representative samples of ore and wet phosphate rocks and beneficiation processes by-products (clay and dolomite rocks, wet screening, magnetic separation and slim) were collected. Natural radionuclides ($^{238}$U, $^{232}$Th, $^{40}$K, $^{210}$Pb, $^{210}$Po) and trace elements (e.g. Cd, Cu, Pb and Zn) were measured using gamma-ray spectrometry, alpha particle spectrometry and ICP-MS analytical techniques. Potential hazards due to beneficiation processes and their by-products were discussed from both the radiological and chemical point of views. Radiologically, internal hazards index, external hazard index, representative level index, gamma absorbed dose rate and occupational dose equivalent due to inhalation were calculated. Chemically, input of toxic heavy metals (radioactive or stable) to the environment due to by-products dump and/or usage were estimated and their hazardous aspects were discussed. Some of the by-products could have potential useful applications such as clay rock and slim for agricultural soil reclamation. The aspects of these useful applications were discussed.

# NORM in clay deposits

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## Abstract

Clay minerals are among the most important minerals used by manufacturing and environmental industries. To reduce the occupational and environmental hazardous impacts of clays' mining, handling and applications, it is essential to investigate their radiological as well as physicochemical characteristics. Twenty one clay's deposit samples from three different regions (Al-Riyadh, Al-Kharge and Jeddah) were collected. Activity concentrations of naturally occurring radioactive materials (NORM), $^{226}$Ra, $^{228}$Ra and $^{40}$K in Bq/kg dry weight were measured using a well-calibrated gamma-ray spectrometer. Radiation dose (e.g. absorbed dose rate- nGy/h and effective dose equivalent- µSv/y), radium equivalent (Raeq) value in Bq/kg and hazardous indices (e.g. external hazardous, internal hazardous and representative gamma level) due to natural radionuclides in clay samples were calculated. The variation of the average activity concentration of natural radionuclides (NOR) according to sampling region could be due to the origin of the geological formation and the geochemical behavior of the NOR. The average activity concentrations of NOR in Saudis' clay samples were much lower than their average concentrations in kaolin clays utilized in Egypt that imported from different countries. Based on calculated hazardous indices (external hazardous and internal hazardous), there are not hazardous effects of clay deposits utilization as a building material. This does not correct based on the average value of representative gamma level that was greater than unity. More detailed studies should be done to consider the occupational exposure due to clay mining and handling.

## Introduction

Clay is a common name for a number of fine-grained, earthy materials. Chemically, clays are hydrous aluminium silicates, ordinarily containing impurities, e.g., potassium, sodium, calcium, magnesium, or iron, in small amounts. Clay deposits vary in the chemical and physical properties and mineral composition, especially in terms of the type of clay minerals dominant. Properties of clay minerals can be determined the field of the use in industry or agriculture. The most common minerals of clay are kaolinite,

montmorillonite - smectite, illite, chlorite, attapulgite or palygroskite, feldspar and calcite. Clay deposits that contain high concentrations of smectite mineral are using for agriculture purposes, because they have desirable properties in agriculture such as a tendency to expand ,disperse, swelling - shrinking behavior as well as high cohesion, also they have a high cation exchange capacity (CEC). While clay deposits that contain kaolinite mineral are using for industrial purposes, because it has less plasticity, cohesion, CEC, and swelling than most other clay minerals [Sheta et al., 2006].

The application of natural clay deposits locally available in Saudi Arabia as soil amended material, soil conditioner, to improve the sandy soil physical condition such as relative swelling, cumulative infiltration, and water conservation were studied. It was concluded that using local abundance clay deposits could improve the predominantly sandy soils in the Middle Eastern countries [Sheta et al., 2006].

Radiation dose due to gamma emitter naturally occurring radionuclides represents the main external source of irradiation of the human body. More specifically, natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in each region in the world [UNSCEAR, 2000].

From the environmental point of view; it is essential to estimate the concentrations of the naturally occurring radioactive materials (NORM) in the clay deposits to ensure their radiological safety for their handling and applications. This study aims at measuring NOR activity concentration in clay samples from three different regions in Saudi Arabia, estimating the radiation doses and possible hazardous impacts of their applications.

## Material and methods

Sampling and samples preparation:

Twenty one clay deposit samples were collected from, three Saudis' regions, Al-Riyadh, Al-Kharg and Jiddah. The selection of the sites was based on previous geological studies [Laurent, 1993]. Sampling was carried out by a scientific group from the soil sciences department, college of food sciences and agriculture, King Saud University. Samples were collected using the standard methods to get a composite sample that represents each site. Samples were dried in an open air of a dry place, mechanically crushed, and sieved through a 2 mm mesh sieve [Sheta et al., 2006].

Gamma-ray spectrometry:

The dried samples were transferred to polyethylene containers of 100 cm3 capacity and sealed at least for 4 weeks to reach secular equilibrium between radium and thorium, and their progenies. $^{226}$Ra ($^{238}$U) series, $^{232}$Th series, $^{40}$K, 137Cs and 210Pb specific activities were measured using well-calibrated gamma spectrometry based on hyper-pure germanium (HpGe) detectors. The HpGe detector had a relative efficiency of $^{40}$% and full width at half maximum (FWHM) of 1.95 keV for 60Co gamma energy line at 1332 keV. The gamma transmissions used for activity calculations are 352.9 (214Pb), 609.3, 1120.3 and 1764.5 keV (214Bi) for $^{226}$Ra ($^{238}$U) series, 338.4, 911.1 and 968.9 keV ($^{228}$Ac) for $^{232}$Th series, 1460.7 keV for $^{40}$K, 661.6 keV for 137Cs and 46.5 keV for $^{210}$Pb. The gamma spectrometers were calibrated using both $^{226}$Ra point source and potassium chloride standard solutions in the same geometry as the samples (Khater, 2001).

**Theoretical calculations:**

Radium equivalent (Raeq) index in Bq/kg is a widely used radiological hazard index and a convenient index to compare the specific activities of samples containing different concentrations of $^{226}Ra$, $^{232}Th$ ($^{228}Ra$) and $^{40}K$. It is defined based on the assumption that 10 Bq/kg of $^{226}Ra$, 7 Bq/kg of $^{232}Th$ and 130 Bq /kg of $^{40}K$ produce the same gamma dose rate. It is calculated using the following equation [Beretka and Mathew, 1985]:

$$Raeq = C_{Ra} + 1.43\ C_{Th} + 0.007\ C_K$$

Where; $C_{Ra}$, $C_{Th}$ and $C_k$ are the activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in Bq/kg, respectively.

The absorbed dose rates due to γ-ray the air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides ($^{226}Ra$, $^{232}Th$ and $^{40}K$) were calculated based on guidelines provided by UNSCEAR (1993, 2000). The conversion factors used to compute absorbed γ -dose rate (D) in air per unit activity concentration in Bq per kg (dry weight) corresponds to 0.462 nGy h−1 for $^{226}Ra$ (of U-series), 0.621 Gy h−1 for $^{232}Th$ and 0.0417 nGy h−1 for $^{40}K$ [UNSCEAR, 2000 & 1993].

$$D = 0.461\ C_{Ra} + 0.623\ C_{Th} + 0.0414\ C_K$$

To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in the air to effective dose (0.7SvGy−1) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) are used. Therefore, the effective dose rate in mSv.y−1 was calculated by the following formula [UNSCEAR, 2000]:

Effective dose rate ($\mu Sv.y^{-1}$) = Dose rate ($nGy.h^{-1}$) X 24 h X 365.25 d X 0.2 (occupancy factor) X 0.7 $Sv.Gy^{-1}$ (conversion coefficient) X $10^{-3}$

According to ICRP (1977) the upper limit of radiation dose arising from building materials is 1.5 $mSv.y^{-1}$ [ICRP, 1977]. For limiting the radiation dose to this value, Krieger (1981) proposed the following conservative model based on infinitely thick walls without windows and doors to serve as a criterion for the calculation of external hazard index Hex - defined as [Krieger, 1981]:

$$H_{ex} = \frac{C_{Ra}}{740} + \frac{C_{Th}}{520} + \frac{C_K}{9620} \leq 1$$

Hewamanna et al. (2001) corrected this model after considering a finite thickness of walls and the existence of windows and doors. Taking these considerations into account, the equation used for the calculation of external hazard index becomes:

$$H_{ex} = \frac{C_{Ra}}{740} + \frac{C_{Th}}{520} + \frac{C_K}{9620} \leq 1$$

The value of this index must be less than unity for the radiation hazard to be negligible, i.e. the radiation exposure due to radioactivity in construction materials must be limited to 1.5 $mSv.y^{-1}$.

In addition to the external irradiation, radon and its short-lived products are also hazardous to the respiratory organs. The internal hazard index (Hin) is used to control the internal exposure to 222Rn and its radioactive progeny. The internal exposure to radon and its daughter products is quantified by the internal hazard index (Hin) which is given by the following equation [Krieger, 1981];

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1$$

For the safe use of a material in the construction of dwellings Hin should be less than unity.

Another radiation hazard index called the representative level index, Iγ, is defined from the following formula [NEA-OECD, 1979; Alam et al., 1999];

$$I\gamma = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \leq 1$$

The safety value for this index is ≤ 1.

## Results and discussion

The statistical summary of activity concentration of naturally occurring radionuclides (NOR), i.e, $^{226}$Ra, $^{228}$Ra and $^{40}$K, and radium equivalent value (Raeq) in Bq/kg dry weight of clay deposit samples was given in table 1,and shown in figures 1 and 2.

There are a noticeable differences in the average activity concentration of $^{226}$Ra, $^{228}$Ra and $^{40}$K, in clay samples from different regions (I:Al-Riyadh, II: Al-Kharg and III: Jeddah). The lowest average activity concentration of both $^{226}$Ra and $^{40}$K, and the highest average activity concentration of $^{228}$Ra were found for Jeddah region samples. Average Raeq values for the three regions samples were comparable and less than 370 Bq/kg. That is equivalent to the maximum permissible limit for indwelling radiation dose due to NOR in building materials.

**Table 1. statistical summary of activity concentration of $^{226}$Ra, $^{228}$Ra and $^{40}$K, and radium equivalent value (Ra-Eq) in Bq/kg dry weight of clay deposit samples**

| Reg. | $^{226}$Ra | $^{228}$Ra | $^{40}$K | Ra -Eq |
|---|---|---|---|---|
| I | 50 ± 3, 7<br>(45-59),6 | 28 ± 3 , 8<br>(16-35), 6 | 823 ± 143, 350<br>(298$^{-1}$181), 6 | 151 ± 17, $^{40}$<br>(92-200) |
| II | 54 ± 11, 31<br>(11$^{-1}$08),8 | 39 ± 5.17, 15<br>(26-69),8 | 781 ± 143, $^{40}$5<br>(262$^{-1}$387),8 | 169 ± 17, 48<br>(72-200) |
| III | 29 ± 7, 14<br>(10-42),4 | 62 ± 13, 26<br>(34-85),4 | 583 ± 104, 209<br>(356- 768),4 | 162 ± 33, 66<br>(86-222) |
| ALL | 47 ± 6, 23<br>(10$^{-1}$08),18 | $^{40}$ ± 5, 20<br>(16- 85), 18 | 751 ± 82, 347<br>(262- 1387),18 | 162 ± 11, 48<br>(72- 222) |

*Average ± standard error, Standard deviation (range), No. of measured samples. I:Al-Riyadh, II: Al-Kharg and III: Jeddah

Average activity concentrations of $^{226}$Ra, $^{228}$Ra and $^{40}$K, and Raeq value for clay samples were 47, $^{40}$ and 751, and 162 Bq/kg, respectively. These values were within the world activity concentration range of $^{226}$Ra, $^{228}$Ra and $^{40}$K in soil; 10-50, 10-50, 100-700 Bq/kg, respectively [UNSCEAR 1988]. There is a trend for the NOR activity concentrations in the different regions where the maximum activity concentration of $^{228}$Ra was found in region III while the minimum average activity concentration of both $^{226}$Ra and $^{40}$K were in region III. This trend is not clear for Raeq value because it is

dependent on the activity concentration of the three radionuclides with weighting factor of 1, 1.43 and 0.07 for Ra, Ra and K, respectively.

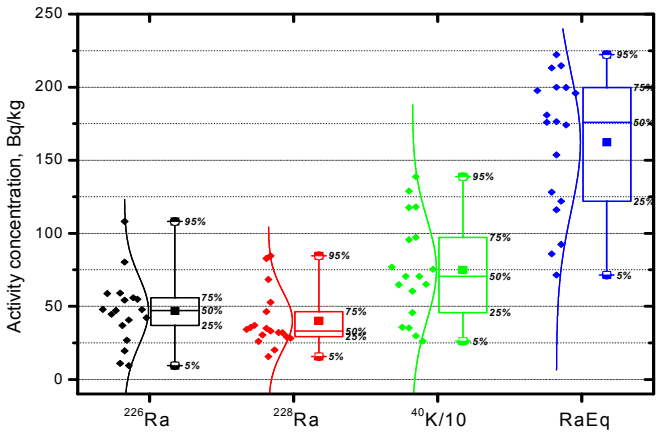


**Fig. 1. Box chart of activity concentration of $^{226}$Ra, $^{228}$Ra and $^{40}$K, and radium equivalent value (Ra-Eq) in Bq/kg dry weight of clay deposit samples.**

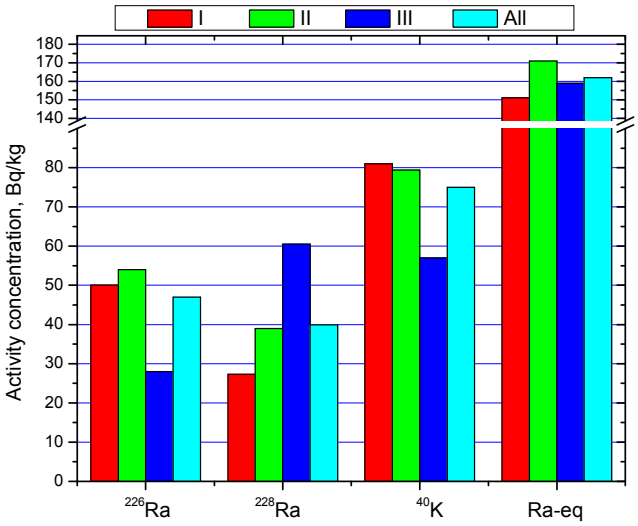


**Fig. 2. activity concentration of $^{226}$Ra, $^{228}$Ra and $^{40}$K, and radium equivalent value (Ra-Eq) in Bq/kg dry weight of clay deposit samples according to sampling region (I:Al-Riyadh, II: Al-Kharg and III: Jeddah).**

Walley El-Dine et al., 2004, studied the activity concentration of NOR in local and imported kaolin (china clay) types used in Egypt. Kaolin is widely used in paper industry, ceramics, refractory bricks, white cement, textiles, rubber, medical industries, and special types of plastics. Their results show an obvious wide range of variation in $^{226}$Ra, $^{228}$Ra and $^{40}$K activity concentrations, and radium equivalent values (Ra-Eq) that had mean values (ranges) of 965 (48-8633), 252 (96-1079), 59 (8-270) and 1329 (188-10185) Bq/kg, respectively [Walley El-Dine et al., 2004].

Clay is a widely distributed, abundant mineral resource of major industrial importance for an enormous variety of uses. Table 2 shows the average activity concentrations of $^{226}$Ra, $^{228}$Ra, $^{40}$K and Raeq in clay bricks as building materials from

12 different countries [Hewamannaa et al., 2001]. The results of Saudi's clay samples are less than the average activity concentrations for $^{226}$Ra, $^{228}$Ra and Raeq, and slightly higher than the average activity concentration for $^{40}$K.

According to IAEA (international Atomic Energy Agency) activity concentration values for exclusion, exemption and clearance (1000 Bq/kg for $^{40}$K and 1000 Bq/kg for any other NOR), it is not recommended that the exposure dose be investigated for the studied clay deposits. Due to the limited number of studied samples and the possible variation in NOR activity concentration, it is recommended to measurement of NOR activity concentrations for more clay deposit samples from the same and other geological formation[IAEA, 2004].

**Table 2. Comparison of activity concentrations and radium equivalents- Raeq (Bq.kg$^{-1}$) in clay bricks in different areas of the world [Hewamannaa et al.; 2001]**

| Country | No. of samples | $^{226}$Ra | $^{232}$Th ($^{228}$Ra) | $^{40}$K | Raeq |
|---|---|---|---|---|---|
| Australia | 25 | 41 | 89 | 681 | 220 |
| China | n.m.[a] | 41 | 52 | 717 | 171 |
| Egypt | 1 | 20 | 14 | 204 | 56 |
| Finland | 33 | 78 | 62 | 962 | 241 |
| Germany | 109 | 59 | 67 | 673 | 207 |
| Greece | 6 | 49 | 24 | 670 | 135 |
| India | 1 | 48 | 52 | 381 | 152 |
| Netherlands | 14 | 39 | 41 | 560 | 141 |
| Norway | 6 | 104 | 62 | 1058 | 276 |
| Sweden | n.m. | 96 | 127 | 962 | 352 |
| Sri Lanka | 24 | 35 | 72 | 585 | 183 |
| Saudi Arabia* | 18 | 47 | $^{40}$ | 751 | 162 |
| Average± stand. Error (range) | - | 55±7 (20$^{-1}$04) | 59±9 (14$^{-1}$27) | 684±70 (204$^{-1}$058) | 191±22 (56-352) |

[a] n.m. indicates that the number of samples was not mentioned in the published work. *Clay deposit samples & Mean ± Stand. Error (range)

Clays and clay minerals occur under a fairly limited range of geologic conditions. The environments of formation include soil horizons, continental and marine sediments, geothermal fields, volcanic deposits, and weathering rock formations. Most clay minerals were formed where rocks are in contact with water, air, or steam. Examples of these situations include weathering boulders on a hillside, sediments on sea or lake bottoms, deeply buried sediments containing pore water, and rocks in contact with water heated by magma (molten rock). All of these environments may cause the formation of clay minerals from preexisting minerals. Extensive alteration of rocks to clay minerals can produce relatively pure clay deposits that are of economic interest (for example, bentonite, primarily montmorillonite, used for drilling mud and clays used in ceramics) [USGS, 1999]. The activity concentration and the environmental behavior of natural occurring radionuclides (NOR) in the geosphere depend on many parameters such as their geochemical properties. Radium (Ra) is an alkaline earth element, and can exist in nature only in the +2 oxidation state. In the pH range of 3 to 10, the uncomplexed ion Ra2+ is the dominant aqueous species for dissolved radium in natural waters. In sulfate- containing waters, precipitation and redissolution of calcium (Ca), strontium (Sr), and barium (Ba) sulfates, rather than adsorption/desorption, could control the concentrations of dissolved radium in the soil. Precipitation of radium is

readily possible as the solid-solution solids (Ba, Ra)SO4 and (metal, Ra)CO3 in water where the concentration of dissolved sulfate and carbonate, respectively, are sufficient high. The adsorption behavior of radium will be very similar to that of strontium. Relative to other alkaline earth elements, radium is the most strongly sorbed by ion exchange on clay minerals. The adsorption of radium is strongly dependent on ionic strength and concentrations of other competing ions that adsorption of radium decrease with increasing ionic strength. Radium is also strongly adsorbed in mineral oxides present in soil, especially at near neutral and alkaline pH conditions. The results of some studies also suggest that radium may be strongly adsorbed by organic material in soils. [EPA, 2004].

The physical characteristics of clay deposits, as one type of sedimentary rock, are governed by many factors of which some include parent rock material, mode of formation of the minerals, the means and distance of transport, and the depositional environment. These characteristics together with uranium, thorium and radium content of the parent rock, and recent physical and chemical events (i.e. chemical leaching, transport with water, and precipitation/adsorption) can affect the final distribution of radium [Edsfeldt, 2001].

Neither radium itself, radium salts, radium carbonates, nor radium oxides are very soluble. However, radium solubility is enhanced by alpha recoil. During the decay of a radionuclide by alpha emission, alpha particles are rejected from the nucleus, carrying off most of the excess energy. The created progeny recoils in the opposite direction. Thus, uranium deposits can kick radium compounds into interstitial pore water due to alpha recoil process [Cothern and Rebers, 1991].

To evaluate the radiation hazardous due to the natural radionuclides in different clay deposits and their various applications, absorbed dose (D) in $nGy.h^{-1}$, effective dose rate (E) in $\mu Sv.y^{-1}$, radioactivity level index ($I\gamma$), external hazard index ($H_{ex}$) and internal hazard index ($H_{in}$) were calculated, Table 3. Calculations of both D and E were considered to estimate the radiation dose due to gamma ray emitter in clay samples. Calculated values are much lower than their value due to natural background (2.4 mSv/y) [UNSCEAR 1993].

**Table 3. statistical* summary of calculated radium equivalent in Bq/kg dry weight of clay deposits sample, absorbed dose (D) in $nGy.h^{-1}$, radioactivity level index ($I\gamma$), external hazard index ($H_{ex}$), internal hazard index($H_{in}$) and effective dose rate (E) in $\mu Sv.y^{-1}$**

| | D | $I_\gamma$ | $H_{ex}$ | $H_{in}$ | E |
|---|---|---|---|---|---|
| I | 75 ± , 21 (44-98) | 1.16 ± 0.14, 0.33 ($0.67^{-1}$.53) | 0.41 ± 0.05, 0.11 (0.25-0.54) | 0.55 ± 0.05, 0.12 (0.38-0.70) | 91 ± 10, 26 (54- 120) |
| II | 81 ± 9, 24 ($33.49^{-1}$06) | 1.26±0.13, 0.37 ($0.53^{-1}$.66) | 0.46 ± 0.05, 0.13 (0.19-0.58) | 0.610 ± 0.07, 0.20 (0.22-0.83) | 100 ± 11, 30 ($41^{-1}$30) |
| III | 76 ± 6, 31 ($^{40}$- 104) | 1.20 ± 0.24, 0.48 ($0.64^{-1}$.63) | 0.43 ± .08, 0.17 (0.23-0.60) | 0.52 ± 0.10, 0.22 (0.24-0.71) | 93 ± 19, 38 ($49^{-1}$28) |
| ALL | 78 ± 6, 23 ($33^{-1}$06) | 1.21 ± 0.09, 0.37 (0.53- 1.66) | 0.44 ± 0.03, 0.13 (0.19-0.60) | 0.56 ± 0.04, 0.17 (0.22-0.83) | 95 ± 7, 29 ($41^{-1}$30) |

**Average ± standard error, Standard deviation (range**

Other calculated indices considered when clay deposits use as a building material where their values should be less than unity. Both internal and external hazardous indices were less than unity that indicates their safe utilization as building material. While, the representative level index values in about 65% of the studied samples and the average value were higher than unity that indicates their unsafe utilization as building material.

## Conclusions

- Average concentration of natural radionuclides (NOR) in clay deposit samples from different regions were varied. That could be due to the different of the geological formation origin and the geochemical behavior of the NOR. Variation of both Ra and K average concentrations were similar (I> II> III) while that of Ra was reversed (III> II> I). Radium equivalent values were comparable in the three regions.
- The activity concentrations of natural radionuclides (NOR) in the studies clay deposit samples are higher than the world average of soil.
- The average activity concentrations of NOR in clay samples were much lower than the NOR concentrations in kaolin clays that imported from different countries and industrially used in Egypt.
- The average activity concentrations of NOR in clay samples were fall in the low range of NOR concentration in clays blocks from different countries that were used as a building material.
- Based on calculated external and internal radiation hazardous indices, the studied clay samples could be utilized safely as a building material.

## Acknowledgement

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# Distribution pattern of NORM on Red Sea shore sediments in relation to non-nuclear industries

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## Abstract

The Red Sea is a deep semi-enclosed and narrow basin that has an intensive non-industrial activities on and near its shore. Oil exploration, phosphate mining and trading, navigation activities and intensive touristic activities are consider as non-nuclear pollution sources that could impose a serious radiological and ecological impacts on the Red Sea marine environment. Both oil and phosphate related activities could increase the concentration of Naturally Occurring Radioactive Materials – NORM such as uranium-238 series, thorium-232 series and potassium-40. Forty representative shore sediment samples were collected from the Egyptian Red Sea shore, from Shuqeir to Marsa Alam City region. Activity concentration of $^{238}U$, $^{232}Th$, $^{40}K$, $^{210}Pb$ and $^{210}Po$ were measured using gamma-ray spectrometry, alpha particle spectrometry and ICP-MS analytical techniques. Previous study showed the possible impact of industrial activities on the activity concentration of NORM in shore sediment. This study will investigate such relation and the distribution pattern of NORM in more details.

# NORM and heavy metals partitioning during water treatment processes

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## Abstract

Water samples were collected from underground water purification plant to study the variation in Naturally Occurring Radioactive Material (NORM) e.g., U, Th and K, and heavy metals e.g., Cd, Pb, Hg, Cu, and Zn concentration through the treatment processes and its relation to physical and chemical properties of water. Samples represent the different treatment processes (input, output, after filtration, sludge tank, reverse osmosis permit and reject, and waste water ponds). NORM and heavy metals concentration in the collected samples were measured using ICP-MS. Their concentrations in water samples show a wide range of variation that depend mainly the water treatment processes and the chemical properties of water samples. Water physical and chemical properties, i.e. pH, EC, major cations (Ca, Mg and K) and major anions ($CO_3$, $HCO_3$, Cl and $SO_4$) were determined. The effect of water treatment processes on NORM concentration; dose assessment due to water drinking (before and after treatment) and the radio-ecological risk assessment were discussed.

# Soil-to-plant transfer factors of $^{210}$Pb and $^{210}$Po in boreal forests

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## Abstract

The general goal in this project was to obtain overview on $^{210}$Po and $^{210}$Pb behaviour and mobility in the environment. Binding and mobility of $^{210}$Po and $^{210}$Pb in forest soil and transfer of polonium and lead from soil to plants were considered. The main study areas were located in Scots pine forests in southern Finland (62º9'N, 22º52'E) and in northern Finland (66º21'N, 26º44'E). The soil samples were collected and separated into different soil horizons (litter, organic and mineral soil layers). Vertical distribution and concentrations of $^{210}$Pb and $^{210}$Po in soils were determined. Wild berry samples and edible mushrooms were collected at the sampling sites. Activity concentrations of $^{210}$Po and $^{210}$Pb were analysed from roots and rhizomes of the berry samples as well as from berries (i.e. fruits), leaves and stems separately. Mushrooms were divided into caps and stipes and each part was analysed separately. The mean $^{210}$Po/$^{210}$Pb ratio was 0.9 in the organic soil layer in the southern and the northern Finland sites. The activity ratios of $^{210}$Po/$^{210}$Pb in the wild berry and mushroom samples were mainly higher than one, indicating elevated concentrations of polonium in the samples. In mushrooms the concentrations of $^{210}$Pb and $^{210}$Po were higher than in fruits of the wild berries. Soil-to-plant transfer factors for lead and polonium will be discussed. The research results gained in this project will enable an assessment of the mobility of $^{210}$Po and $^{210}$Pb in the environment and in the food chains and estimation of ensuing radiation doses to humans.

# Radiological assay techniques associated with TENORM industry

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## Abstract

In recent years there has been an increasing awareness of the radiological impact of non-nuclear industries that extract and/or process ores and minerals containing naturally occurring radioactive materials (NORM). Actions were undertaken to find and characterize the main industrial activities involving NORM and to assess the impact of these activities on the nearby public. The main concern was on past activities in phosphate fertilizers industry and the associated by-products disposal as well as the activities in oil and gas production and processing industry. Phosphogypsum is the main by-product generated during the phosphoric acid production process (wet process), and phosphate slag is the principal by-product generated from the production of elemental phosphorus (thermal process). During the wet process, a selective separation and concentration occurs for radionuclides which are naturally associated with the phosphate ores. In oil and gas production and processing equipment some elevated TENORM contamination might occur due to the physical processes to which the extraction fluids are exposed. Some radium and radium daughter compounds are slightly soluble in water and may become mobilized when this production water is brought to the surface. The paper presents the methodology used in some particular cases to deal with NORM characterization as a regulatory requirement for non-nuclear practitioners to improve their radiological safety and to achieve an adequate level of knowledge as concerns the risks associated with NORM industry.

# NORM management in the oil & gas industry – Saudi Aramco experience

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## Abstract

It has been established that Naturally Occurring Radioactive Materials (NORM) may accumulate at various locations along the oil/gas production process. Components such as wellheads, separation vessels, pumps, and other processing equipment can become NORM contaminated, and NORM can accumulate in the form of sludge, scale, scrapings and other waste media. This can create a potential radiation hazard to workers, general public and the environment if certain controls are not established. Saudi Aramco has developed NORM management guidelines and is implementing a comprehensive strategy to address all aspects of NORM management which aim towards enhancing: NORM monitoring, Control of NORM contaminated equipment, Control of NORM waste and disposal, Workers protection, awareness, and training The benefits of shared knowledge, best practice and, experience across the oil & gas industry are seen as key to the establishment of common guidance. This paper presentation outlines Saudi Aramco's experience in the development of a NORM management strategy and its goals of establishing common guidance throughout the oil and gas industry.

# The needs and feasibility of land reclamation of areas affected by enhanced natural radioactivity

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## Abstract

The major efforts devoted to disposal of radioactive materials are focused on those originating from nuclear industry. Far less attention has been paid to environmental burden of waste with natural radioactivity enhanced by non-nuclear industry. Such waste differs significantly from classical nuclear ones and the radiation risk is often associated with risk caused by other pollutants. Contrary to the nuclear waste that's strictly controlled, it has been a common practice to put TENORM *(Technologically Enhanced Naturally Occurring Radioactive Material)* into heaps, where they can reach thousands of cube meters or tonnes, without any protection. Exposure to meteorological conditions sets some chemical or physical processes in motion, leading to the selective transfer and accumulation of radionuclides and disequilibrium in decay series.

As a result of the inconclusive European law, that left the decision where risk caused by TENORM is significant under each member state's competences, non-nuclear industry is hardly ever aware of environmental problems caused by natural radioactivity or expect negative consequences in case of implementing radiation protection measures. This results in the substantial underestimation of the detrimental effects to the environment originating from TENORM. EU member states apart each other try to regulate this problem case by case when but the risk caused by TENORM is rarely taken into account when the treatment of such waste is planned.

The feasibility of different methods of land reclamation was discussed including hard land reclamation, bioremediation and phytotechnology. An approach based on dilution of TENORM with inert materials was considered. Contrary to the nuclear waste, where dilution is totally forbidden, natural radioactivity, which is present in small amounts elsewhere, is a case where such approach to decrease risk sounds rationally. In extraction industry where huge amount of gangue is present, such approach is well-founded also from economic point of view.

## Introduction

Natural radioactivity is ubiquitous in human environment. According to the state-of-the-art radioprotection, radiation emitted by primordial radionuclides in their natural state that has not been altered due to human activity is not considered to be a source of harmful effects neither for human beings nor environment. There are many areas in the

world having elevated so called "natural background" caused either by the geological and geochemical structure of the rocks, or by the radioactive content of water flowing from underground springs. Whether or not it can cause a negative or positive effect on human beings is a matter of opinion. But if concentrations of natural radionuclides have been changed as deliberate or accidental action carried out by human being it is quite another matter. The classical case where radiation risk caused by natural radioactivity is not negligible is uranium mining and milling. It is abundantly clear that such processes must be carried out at the region where reach uranium ore occurs. But such activity is considered to be an immanent part of nuclear industry to be enclosed in radiation protection domain at the very beginning. After the enhanced natural radioactivity had been thoroughly studied in other industries it became clear that such phenomena are very frequently present in the anthropogenic environment. Many processes beyond nuclear industry lead to a situation where the activity concentration of naturally occurring radionuclides is enhanced. Such alteration of natural state can result in increased radiation risk to the people as well as to environment and non-human biota. Hence, the monitoring and prevention of occupational radiation risks caused by enhanced natural radioactivity has become obligatory in many cases of industry of concern.

Enhanced natural radioactivity is usually associated with industrial processes where a significant mass reduction of raw materials occurs. As a matter of fact, these processes are not aimed at production of natural radionuclides or the deliberate use of radiation. Therefore, radioactive nuclides are often accumulated in useless industrial residues. From the point of view of the general principles of radiation protection, the activity concentration of natural radionuclides in such materials is sometimes high enough to rank them as radioactive waste. Their amount collected in one site frequently riches hundreds of thousands of cubic meters or tonnes. For instance, in coal mining industry radium activity deposited in single tailing ponds may reach as high as 300 GBq (Michalik et al. 2005). Probably, the biggest "producer" of waste with enhanced concentration of natural radionuclides are phosphate processing plants where radionuclides remain associated with the phosphogypsum particles, being subsequently stored in a disposal sites located in the vicinity of the factories with surface dose rate reaching 350 MBq/h (Bolivar et al. 2009). In spite of that TENORM-type waste (TTW) is often deposited directly into the environment, what is strictly forbidden in case of "real" radioactive waste. Contrary to the occupational risk, which is more or less controlled, by far, less attention has been paid to the environmental burden emerging due to TENORM-type waste (TTW). This results in sites where such waste have been dumped not to undergone adequate land reclamation and often even monitoring of radioactive pollution is not carried out. There are only few examples where TENORM type waste are treated in correct way (Welbergen and Wiegers 2008). There are significant number of cases where only non-radiological parameters are effectively taken into account during presumptive land reclamation, mainly due to the lack of proper regulation.

## Environmental burden of TENORM residues

There is a lot of data dealing with the behaviour of natural radionuclides being in the natural state on the border of abiotic and biotic matter. Frequently, the processes of metabolism lead to concentration of some long-lived natural radionuclides in particular tissues of fungi, plants as well as animals (McDonald et al. 1996). Derived committed dose can be higher than doses resulted from artificial radionuclides accumulated simultaneously (Aarkrog et al. 1997). There are some examples of societies based on limited trophic chain where the related committed dose to individuals caused by biologically accumulated natural radionuclides such as polonium and lead ingestion is significant. J. Van Oostdam et al. (2005) indicated $^{210}$Po derived doses as high as 10 mSv per year for some aboriginal northern communities consuming large amounts of caribou. Either, annual doses reaching 3 mSv were reported for population of fisherman just living on seafood (Alonso-Hernandez et al. 2002), (Camplin et all. 1996). If such processes are going on in unchanged environment, one can easily image what is going to happen in the vicinity waste dump where residues containing enhanced concentration of natural radionuclides had been collected.

Each particular occurrence of TTW presents a unique scenario of exposure – usually different from those caused by artificial radionuclides present in radioactive waste or spent nuclear fuel. As a result of the direct contact with environment, some transformation processes such as mobilisation of radionuclide species from solid phases or interactions of mobile and reactive radionuclide species with components in soils and sediments may be set in motion (Vandenhove and Van Hees 2007). Also considerable transfer of radionuclides to biota can be observed (Soudek et al. 2007). All these result in that the original distribution of radionuclides deposited in environment can change over time. Moreover, natural radionuclides are often associated with other pollutants as heavy metals or hydrocarbons that can escalate negative impact on environment if dumped out of plants. That's why, to plan the land reclamation of a site affected by TTW, information on radionuclide species deposited, interactions within affected site either local ecosystems or environment compartment and the varying in time distribution of radionuclide species influencing mobility and biological uptake is essential (Tamponnet et al. 2008).

## TENORM-type waste characterization

TTW produced by non-nuclear industrial activities such as mineral production, mining activities or coal fired power plants contain a number of long-lived natural occurring radionuclides from the uranium and thorium decay series. The main, from radiation protection point of view, properties of TTW which make them significantly different from "classical" radioactive waste are (Michalik 2007);

- the occurrence in bulk quantities deposited directly in the environment,
- the wide variety of radionuclides speciation and different minerals content,
- the possible coexistence of other pollutants as heavy metals, sulphates, hydrocarbons

Taking into consideration the radionuclides that occur in TTW and either their activity concentration or total activity, some part of them should be classified as radioactive

waste containing alpha emitters, (i.e. the limit of activity concentration for radium isotopes is 10 kBq/kg). Actually, the decision to rate TTW among the radioactive waste at all, not only as alpha emitters is rarely taken. It results in technical problems and economical consequences that would follow such decision (to remind, such waste must be sealed and deposited in a repository, in case of alpha emitters in deep underground repository). After they are classified as radioactive waste TTW would generate enormous cost- related implications and fill available repository very quickly. There is no possibility to treat them as such with regard to existing regulation, which had been prepared when thinking about nuclear waste. Finally, there are gaps, no special regulation and no correct way of deposition. Moreover, from environmental point of view, very interesting is, what one should do with waste that contain slightly lower than limit radioactivity content, in case of radium, for example, 9 kBq/kg. Expected effects on environment caused by waste with 10 and 9 kBq/kg of radium should not be significantly different. Actually, the proposed clearance level for natural radionuclides from uranium and thorium decay series have been set at the level of 1 kBq/kg. So, even if one wants to follow the existing regulation quickly will meet the big gap between 1 and 10 kBq/kg.

After deposition TTW in environment, enhanced concentration of natural radionuclides first of all, results in enhanced exposure of biota to external gamma radiation. So, accurate measure of the total radionuclides concentration in these waste materials is crucial to asses the potential radiological risk at a dump site. However, if one managed to gather data about physical presence of each particular radionuclide, this information would give only a part of the knowledge necessary to evaluate its harmful potential. The radiological hazards can be increased by migration process of mobile fraction of these radionuclides to the vicinity of a depository (Sheppard et al. 2005). Being released into an ecosystem, they can enhance the gamma radiation doses to biota. That is the reason why it is crucial to know the mobility of radionuclides (Martinez-Aguirre et al. 1995). The mobility and environmental behaviour of every element depend on their speciation in certain waste material (Zhongwen et al. 2002). The speciation of a radionuclide is generally related to its physical and chemical forms existing, that is, simple and complex ions in interstitial solution, exchangeable ions associated with waste material organic fractions occluded or co-precipitated with metal oxides, carbonates, sulphates and other secondary minerals.

But the exposure to external gamma radiation is only a tip of an iceberg. One should remember that in thorium and uranium decay series there are 7 and 12 alpha particles respectively (additional 12 are in actinium series). Even when one consider decay series starting from radium ( $^{226}$Ra or $^{228}$Ra ) what is very common in TTW waste the number of alphas decreases only to 7 and 9, respectively. It is hardly ever taken into consideration but in environment, in case of plants, especially plant roots, the exposure to external alpha radiation is as important as exposure to alpha radiation emitted by incorporated emitters. Also one should remember about betas emitted by natural radionuclides. Actually the weighting factors for alpha and beta radiation established for human being can not be directly applied for plants but there are no rational reasons why they should be significantly lower. Direct measurements showed that absorbed dose resulted from alpha radiation can reach the same level as doses from gamma radiation (Michalik 2008).

Besides the problems caused by activity concentration, radionuclides speciation and migration, the evolution of the related risk must be taken into consideration. Namely, the most common radionuclides responsible for risk creation are radium isotopes. In case where source of TTW are formation waters (oil and gas industry, underground mining) radium isotopes, both from uranium and thorium series, $^{226}$Ra and $^{228}$Ra are dominant. It means that, significant disequilibrium in natural decay series exists. At the beginning, just after deposition, there are almost pure radium as the only contaminant (besides possible other, non radioactive ones, of course). Radium isotopes are usually weakly mobile and not bio-available in environment. As it was proven radium creates not soluble radium-barium sulphate or its atoms are strongly bonded with fine clay minerals. The bio-available part usually does not exceed 1 % of total activity (Leopold et al. 2007). So, the environmental risk is limited only to the exposure to external gamma radiation, or as it was shown above, alpha radiation and finally could be partially limited by a cover from an inert material. Moreover, the part of exposure derived from radium $^{228}$Ra will relatively quickly decreases (the half live of $^{228}$Ra is 5.7 years, so not supported by long-lived parent radionuclides $^{228}$Ra will quickly disappear). The only difficult situation is that, in the decay series started with not-supported radium $^{228}$Ra, the activity concentration of decay products i.e. $^{224}$Ra can exceed the activity concentration of radium after few years. It can cause troubles during measurements and appropriate actual dose assessment.

Quite different situation exists in case of radium $^{226}$Ra. It decays slowly. Not only from the point of view of a human being but also form point of view of the environment it lasts eternally. 1600 years is long enough to be eye witness of changes going on in an ecosystem.

As it was already mentioned, the bio-availability of radium is very weak, but such comfortable situation with immobilised radium does not last long. Taking into consideration the relationships inside the uranium decay chain one should expect not so quickly, but permanent growth of long lived radium daughters as lead $^{210}$Pb and polonium $^{210}$Po. Both these radionuclides are very well known as easy migrating nuclides in environment and available to biota. After decay, the nuclide of radium is released from the barium-sulphate or clay mineral cage. Moreover, between radium and lead and polonium, there is radon (or thoron respectively) occurring in gaseous form. So, after one hundred years one should expect in the site where TTW with radium had been dumped exactly the same activity of easy migrating and easy bio-available lead and polonium isotopes. Besides specific chemical properties, they are beta and alpha emitters respectively and both of them are chemiotoxic elements.

In summary, the proper land reclamation of sites affected by TTW is an interesting challenge.

## Remediation trials

Among noticed approaches to the reclamation of land affected by TTW, the simplest one, compliant with radiation protection rules, is to treat such waste literally as radioactive ones and apply all required restrictions. But for many reasons, it is applicable in limited cases (Michalik 2008,2). Usually, only parts of dewatering systems from crude oil and natural gas industry (Varskog 2007) or gangue from mineral sand processing (Hutchinson and Toussaint 1998) are disposed to especially

prepared repositories. Sediments had been created in surface tailings sometimes are treated in special way (Al-Masri and Suman 2003) but usually, they are left without any action. Some promising efforts to wash out radium from oil sludge were made in Egypt (Afifi et al. 2009). Also applications of biotechnology, in order to make radium more mobile from uranium milling waste were noticed (Muñoz et al. 1995). But any of them have been applied in technical scale. In uranium mining and milling industry, where implementation of strict rules of radiation protection has long history, usually a kind of hard remediation is applied. Land-filing and covering with an inert matter is sufficient to limit the exposure to external radiation as well as further radon exhalation (Krizman 1995), (Juhász et al. 2001). But this approach is not sufficient enough to stop radium and further polonium migration.

The possibilities of application of different methods of land reclamation have been considered towards the sites contaminated by radium rich sediments originating from underground coal mining (Chałupnik et al. 2001), (Michalik 2004). The first based on application of phytotechnology was tested on an abandoned mining settling pond. Six-year-lasting observation of contaminated area let one noticed that the process of natural plants transgression was so effective and, even without any support, good enough to stop the physical propagation of contamination. The plants overgrowing the pond created a tight cover able to stop water and air land erosion. It supposes that, in case of controlled and supported propagation of selected plant species, it would be actually an effective and cheap method for immediate land reclamation of contaminated with radium sites.

The possibility of phytoextraction at this area was evaluated for two plant species *Cirsium vulgare* and *Calamagrostis epigejos*. The balance of radium $^{226}$Ra in plant and sediment at the tested areas showed that one can expect only less than 0.01% of total amount of radium will be extracted during one vegetation season (Michalik et al. 2009). So it is by far too small for effective application of this method. Moreover, the experiment was done based on sediments in which radium was mainly adsorbed at clay minerals. Other experiments, done on sediments with radium-barium sulphate, a hardly soluble mineral, let one to expect even lower results. So, such approach does not solve the problem at all.

The recommended and usually followed approach to utilisation of sediments that had been gathered in underground workings is to put them into old galleries considered never been used again. From the radiation protection point of view such approach is optimal for safe disposal of sediments from surface settling ponds too. However, from technical point of view a lot of obstacles exist as limited capacity of empty underground spaces or distance from settling pond to the nearest shaft in use.

Finally, an approach based on dilution of radium waste was taken into consideration. In general, dilution of radioactive waste is totally forbidden. But in case of natural radioactivity, which is present in small amounts elsewhere, such approach to decrease derived risk sounds rationally. Especially in case of hard coal mining, where technological process creates fovorable circumstances to do it in economic way. During coal exploitation process, at least, half of mine spoil is waste rock and gangue. The radium activity concentration in thse waste do not differ significantly from average value taken as background for earth crust. So that it would be a good “solvent” for radioactive sediment. Actually, mechanical mixing of huge amount of mineral waste is

complicated and expensive but again, coal exploitation process provides an opportunity to do it as a "by the way". Namely, all excavated matter must pass through a coal treatment plant in order to clean coal from gangue and prepare expected fraction of coal by flotation. The total amount of flotation and coal cleaning waste is big enough to turn back radium activity concentration in sediments to background level after homogenous mixing.

The possibility to apply such approach was tested in a mine. The total amount of mine spoil: coal and all types of created waste were balanced against total amount of radioactive sediments gathered in water galleries at all mine levels. It have been done year-by-year since 2005 (table 1). Codes were given based on rules of European waste catalogue (Michalik 2009).

**Table 1. The balance of the excavation process.**

| year | Mine productivity (t): | | | | | |
|---|---|---|---|---|---|---|
| | total | coal | Waste from coal cleaning process code: 010102 | Waste from flotation code: 010481 | Sediment from water galleries code: 190899 | Sum of : 010202 and 010481 |
| 2005 | 6895262 | 3674000 | 3050585 | 170677 | 1740 | 3221262 |
| 2006 | 6760336 | 3703900 | 2894547 | 161889 | 1620 | 3056436 |
| 2007 | 7056571 | 3737600 | 3163933 | 155038 | 3668 | 3318971 |

The current and archive data concerning radium activity concentration in every kind of considered materials were used (table 2). Because there are no data about behaviour of such sediment during coal enrichment process, all possibilities were taken into account, it means: total amount of radium accumulated in particular kind of waste have been considered separately (table 3).

**Table 2. Basic statistics of radioactivity in sediments and gangue.**

| | code: 190899 | | code: 010202 & 010481 | |
|---|---|---|---|---|
| | Ra-226 | Ra-228 | Ra-226 | Ra-228 |
| | Bq/kg | | | |
| average | 705,6 | 364,4 | 79,7 | 73,1 |
| median | 409,0 | 246,0 | 73,0 | 79,0 |
| Minimum | 21,2 | 19,0 | 24 | 10 |
| Maximum | 8272,0 | 2880,0 | 189 | 112 |
| Number of samples | 39 | | 15 | |

**Table 3. Radium activity concentration in mixed waste.**

| nuclide | year | 190899 + 010202 + 010481 | | 190899 + 010102 | | 190899 + 010481 | |
|---|---|---|---|---|---|---|---|
| | | *maximum* | *average* | *maximum* | *average* | *maximum* | *average* |
| Ra-226 [Bq/kg] | 2005 | 193,36 | 80,07 | 193,61 | 80,09 | 270,57 | 86,05 |
| | 2006 | 193,28 | 80,06 | 193,52 | 80,08 | 269,08 | 85,93 |
| | 2007 | 197,92 | 80,42 | 198,36 | 80,45 | 375,81 | 94,19 |
| Ra-228 [Bq/kg] | 2005 | 113,49 | 73,22 | 113,58 | 73,23 | 139,93 | 76,01 |
| | 2006 | 113,47 | 73,22 | 113,55 | 73,23 | 139,42 | 75,95 |
| | 2007 | 115,06 | 73,39 | 115,21 | 73,40 | 175,97 | 79,80 |

Obtained results seem promising. The average activity concentrations in all end-products of the process are slightly increased in comparison to their original value. Moreover, considering the worst case scenario, it means the total amount of radium remains in the flotation waste, the smallest contributor to the total mass of concern, the activity concentration does not differ significantly from other ones and there are no limitations in their disposal at surface mine spoil bank and further use as i.e. aggregate.

## Conclusion

In the light of different approaches to remediation of areas affected by waste with enhanced concentration of natural radionuclides, the dilution method with inert material or waste originating from industry of concern seems to be well justified from technical and economical point of view. The example from coal mining industry shows, based on the balance of waste rock and gangue produced by every mine, that there are enough capabilities to use this technology for safe disposals of radium-rich sediments that had been gathered in surface settling pond due to either former or current mining activity. However, such approach needs to be approved by appropriate regulation.

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# Uranium and heavy metals in narghile (shisha, hookah) moassel

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## Abstract

Cigarette smoke is a source of some trace and heavy metals but also the smoking mixtures used in narghile. Some cigarette filters retain significant quantities of cadmium, lead, magnesium, iron, and other metals. However, data on narghile (hookah, shisha, water-pipe) are scarce. So, the objective of the present study was to investigate the contents of the main and widely used smoking product: moassel. Ten representative samples of 3 different moassel brands were collected from the local market of Cairo city-Egypt and Riyadh City- Saudi Arabia. Uranium and heavy metals (e.g., Cd, Cu, Pb and Zn) were assessed using ICP-MS. Among the 10 representative samples of the 3 different moassel brands, The results indicate the existence of a wide range of variations in uranium and other element concentrations that could be because of non standard manufacture processes. The concentration levels were compared to the results of other studies and that of cigarette tobacco. Our study shows that, as far as trace elements are concerned, harm can be reduced. Public health officials could include in the national prevention plans the use of smokeless tobacco, particularly when addressing heavy narghile smokers. However, it must be clear that there are important differences between smokeless products. On one hand, some of them, like the Sudanese “tumbak”, contain high levels of carcinogenic nitrosamines and we fear that Arabian shamma might be of a similar nature. On the other hand, a moist snuff like the Swedish SNUS, is, in the view of prominent international experts, highly recommendable all the more that it is also very low in carcinogenic substances. We therefore encourage its use all the more that this harm reduction product is culturally adapted to the Arab world context and other similarly sanitary and socio-cultural ones.

# External gamma radiation produced by materials: proposal of an evaluation model. Application case study NORM

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## Abstract
External gamma exposure to natural radiations is due to a series of isotopes gamma emitting, contained in all building materials. Beginning from the set up of a calculation model to estimate the emission in a closed environment, it was possible to derive the effective dose in the environment. The case of materials generally rich of natural radioisotopes was investigated.

From an inverse process, starting from the examination of the radiation field it is possible to estimate the extension of an activated surface. A case of contamination and activation surface in a general context as the inner part of nuclear plant or a radioactive facilities was evaluated. This method, moreover, is also applicable to NORM.

## Introduction
The emission of total gamma radiation of simple or variegated structural configurations does not represent a simple modeling phenomenon, since the factors associated with emission, absorption and reflection, principally due to radiation-material interaction, generate a series of secondary events, the precise estimation of which inevitably risks complicating the problem.

Emitted radiation comes into being, due to various processes, involved in a series of intermediary interaction, then to be definitively absorbed. A global balance can be created considering the equation.

$$\text{Incident Radiation Energy} \rightarrow \text{Converted} + \text{Absorbed Radiation Energy} \qquad (1)$$

At this stage, it becomes inevitable to refer to the “medium”, or rather to the space-material reference in which the radiation is to be found.
Introducing a medium in which radiation is propagated, has the substantial aim, not just of identifying parameters such as propagation speed or refraction index in the medium, but of verifying if such a medium is of influence in terms of radiation intensity reduction, and therefore of absorption - bremsstrahlung of the same.

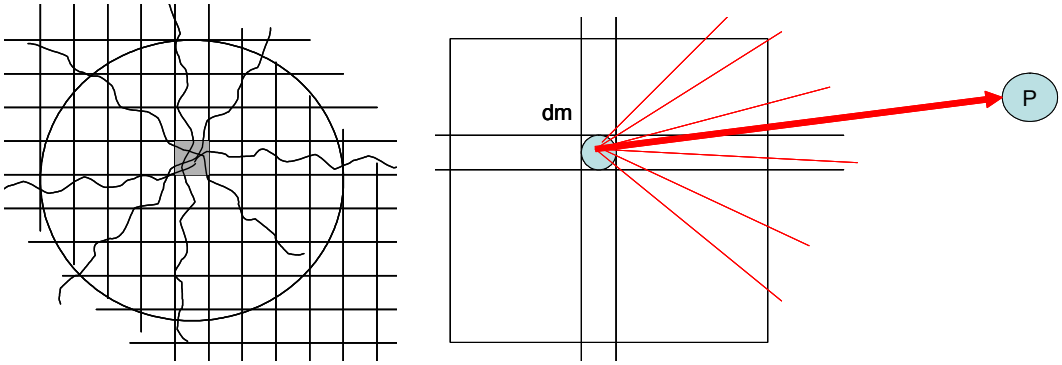


**Fig. 1. Finite radiant element in the material matrix (left), and activity produced by a finite element on a P external point.**

The study conductible on the emission of a finite source will start from a global approach, considering the attenuation properties of the finite elements and the data inherent to the absorption properties[1] of the interested medium being available. In the treatment of the subject, which will be carried out, the material presents prevalent gamma emissions (radionuclides greatly present), uniformly distributed and unequivocally determined by spectral lines. It is possible to hypothesise in order to reduce a sample in finite elements, considered as precise sources for simpler treatment, yet it is computationally suitable at the same time.

In the majority of cases, the level of activity of a source varies by varying the distance of the P observation point from the source, and from the properties of the medium crossed by ionised radiation.

$$A(P)=A(Ao, \underline{S}, \rho) \quad (2)$$

where:
Ao = activity in correspondence with the emission point;
P= observation point or radiation exposure target;
$\underline{S}$ = vectorial coordinate of point P compared to source origin;
ρ = specific density of material seen in relation to its mass attenuation;

## Theory

The final objective of this study of materials is to determine the dosimetrical units associated with exposure to emitted radiation. The knowledge of the dose values descends directly from the values of the field typical parameters. In such a paragraph Laws are presented in order to determine the flow density for definite sources in space, in the hypothesis that the interplaced medium is air and that the point-source distance is such that it is able to disregard the effects of secondary absorption.

It is known that an isotropic point source is for the simplest case of emission. Photons are uniformly emitted in all the $4\pi$ solid angle, and in the absence of absorption, the φ flow density results as being thus defined (Pelliccioni, 1993):

$$\varphi = \frac{S}{4\pi r^2} \quad (3)$$

[1] Each medium considered has known properties such as density and degree of radiation absorption. Moreover the spectrometric analysis add information relative to the emissive properties of the material. The latter data can be evaluate according to an approach which takes into account each single radionuclide and of its own emission frequency, or rather, its total radiation contribution given by the finite material block.

At this point, also the attenuation produced by auto-absorption of the same materials comes into play, as well as the air. It is necessary to introduce a further complication (Salinas et al., 2006). Returning to the formula for photon attenuation, the pass-ratio results as being:

$$f = 1 - \frac{N}{N_0} = 1 - e^{-(\eta/\rho)\,\rho l} \tag{4}$$

For mono-energetic radiation the mass attenuation coefficient is known, as is the density of the material crossed. The term "l" represents the distance in the material with which the equi-balanced source sees the target[2]. The latter being dependant on the spatial coordinates of the integration dominion (Alitto, 2005), even the f function will depend on them.

$$\varphi = \iiint_V \frac{S_v \cdot f(x,y,z)}{4\pi r^2}\,dx\,dy\,dz \tag{5}$$

The study of an almost standard indoor environment with six walls, is conducted by the study of each individual wall. For exposure symmetry the maximum emission plane is situated along the axis of each wall. Radioactive emission of a wall presents a bell trend with the maximum situated in correspondence with the centre axis of the wall. Since gamma radiation covers a certain distance in the air ($1.2 \cdot 10^4$cm) it is a reasonable hypothesis to consider that the emissions of each wall are overlapped and culminate in the central area of the environment where people are more likely to be residing.

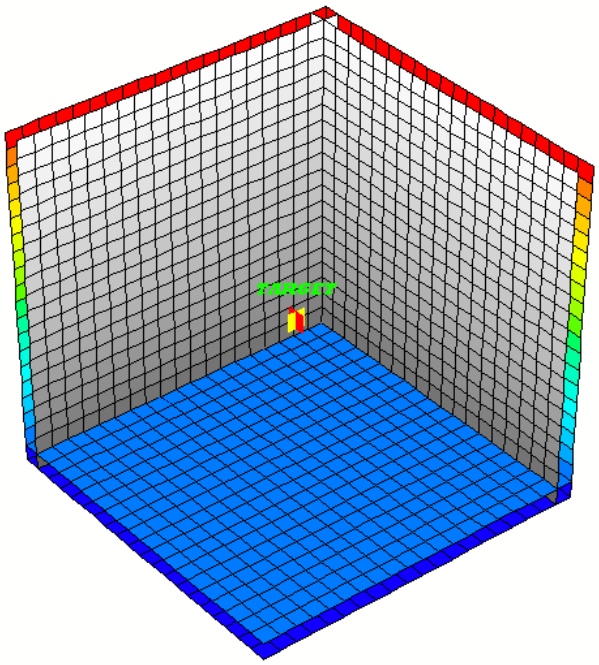


**Fig. 2. Identification of the calculation target of a hemi – environment.**

[2] Supposing that the wall is on an xy plane, that the target has P(x0,y0,z0) coordinates, that the distance of P from the wall is d, r the distance of a generic radiant point dV of the wall from P, then the distance $l = r\left(1 - d/\left|z - z_0\right|\right)$.

Furthermore, considering the absorption power of the air is translated into the introduction of a multiplicative factor similar to f(x,y,z), considered with the parameters of standard air composition.

$$\varphi = \iiint_V \frac{S_v \cdot f(x,y,z)\ f_a(x,y,z)}{4\pi r^2}\,dx\,dy\,dz \qquad (6)$$

Such a further contribution leads to the natural extension of the formula to the case in which, in the virtual path of radiation, from the source to the target, a series of different mediums and materials, components such as mixed-layer thicknesses with different attenuation properties (Stranden, 1979), is in existence.

$$\varphi = \iiint_V \frac{S_v \prod_i f_i(x,y,z)}{4\pi r^2}\,dx\,dy\,dz \qquad (7)$$

Nevertheless, the lack of "good geometry" conditions leads to the necessity of taking into account the build-up effect (Brar et al., 1999), on the basis of the components which characterize the materials. A corrective factor will be correlated to the formula (7) which is equal to

$$B(E,r) = 1 + \frac{(b-1)(K^x - 1)}{K-1} \qquad (8)$$

The formula to undergo simulation will be the result of the following mix (Alitto,2005):

$$\varphi_{sim} = \frac{S_v}{4\pi} \iiint_V B(E, r(x,y,x)) \frac{\prod_i f_i(r_{(x,y,z)})}{r^2}\,dx\,dy\,dz \qquad (9)$$

Validation of the simulations was carried out using a comparison of the results with scenarios that have already been experimented and calibrated (R.P. 112). By means of the data provided by it, it was possible to carry out the direct identification of the external gamma dose, due to the presence of walls, floors, surfaces, thicknesses etc., by means of the conversion formula:

$$D^i_{\gamma\text{ - ext}} = \varphi \frac{c_i}{S_V} \qquad (10)$$

Since the valuation of the external dose in mSv/year is through the knowledge of the fluence rate φ, obtained from the simulation, from the typical mass photon flow of the material and from $c_i$[3] which is a characteristic function of the i single radionuclide considered, in the case of mono-energetic radiation. The extension of poly-energetic radiation (Higgy et al.,2000), therefore a classical distribution of nuclides in the material matrix, is obtainable simply as a summary of the contributions of the single nuclide dose.

[3] The term Ci depends on the concentration of activity of the natural gamma-emitter element at a given frequency, and by a equal scale factor characteristic of the considered standard environment in R.P. 112 for 4x5x2,8 m³ , with wall thickness equal to 0,2 m and radiant material uniformity conditions.

## Materials and methods

The field of radiation produced by a single finite wall, in which a certain concentration of natural radionuclides results as being uniformly distributed, can be inferred through the use of the equations seen previously, with the use of an appropriate calculator, through the use of volumetric numerical resolution, through libraries which are, in part, present in the software used (Matlab 7.0).

Such graphic representations of the emitting wall allow for the hypothesis that, in a preliminary and evidently superficial way, the previously formulated theory is correct, or rather that the field produced in a standard indoor environment (6 walls, of which two floors and four horizontals) is at its greatest in the central point.

The incorrectness of this assumption was verified proceeding with a volumetric analysis of the volume considered tridimensionally.

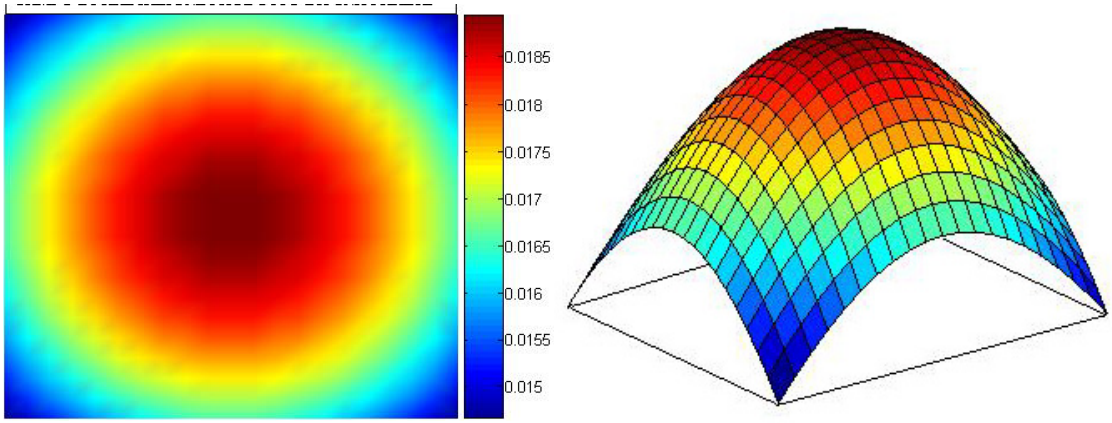


**Fig. 3. Radiation field produced by a single wall by materials containing natural gamma emitters per unit of mass photon flow (left) and graphic visualization of the field produced by a wall as 3D surface (right).**

In the first phase, the distribution of a reference mono-energetic source (K-40), uniform in all the material forming the 6 walls of a standard indoor environment, having cubic dimensions (4 x 4 x 4 $m^3$), and equal thickness for each wall (0,2 m) was considered (Alitto, 2005).

A tridimensional calculation model was created which considered, for the evaluation of the field of radiation, volumetric elements of the room that were sufficiently small to be able to consider, the reasonably constant field of radiation within them.

The centres of each of these volume elements, aligned according to the three spatial directions, form the calculation plane of the field of radiation, or rather the planar portions of the environment space, to evaluate the variability of the field of radiation.

## Results and discussion

The simulation was carried out, taking into consideration the aforementioned parameters, for the determination of the fluence rate of gamma radiation in each point-packet of the volume delimited by the walls according to the schematisation highlighted in the following image.

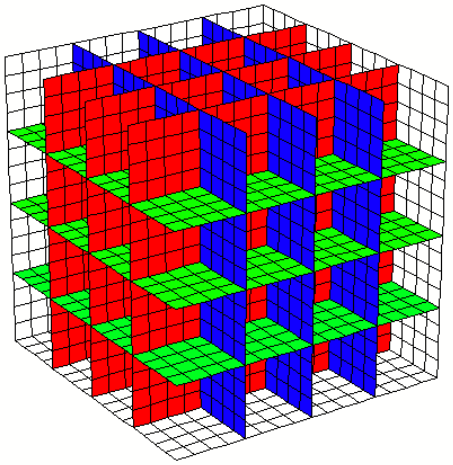

**Fig. 4. The hemi-environment delimited by the walls (white grid), and the calculation planes (coloured) along the three directions. Some of them are shown as examples.**

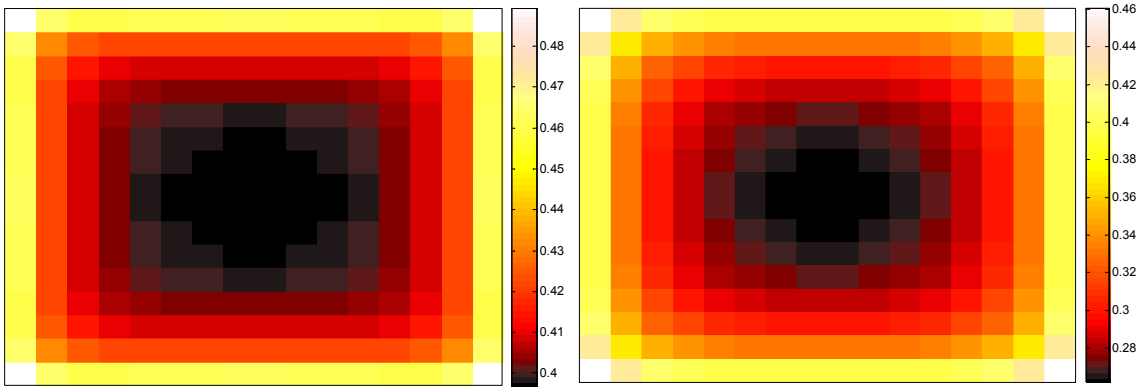


**Fig. 5. Colorimetric evaluation of the field of gamma radiation induced by a source distributed per unit of photon volume flow, in proximity to one of the radiant walls (left) and for a central section (right).**

The symmetry intentionally chosen for the case, which does not preclude the possibility of extending the model to scalene dimensional parameters, allows for the carrying out of an exhaustive evaluation of the calculation planes once for all components, taking one of them as a starting point.

From the compared images it is possible to note how, in reality, the hypothesis carried out previously is not correct. The most intense points of the field of radiation are represented by the areas contiguous to the walls. In substance, the greater effect of the field is in the vicinities of the corners. Instead, the centre of the environment must be considered as that having the lesser radioactive impact.

The example was executed in the maximum symmetry conditions: in this case, the reference environment was cubic, with uniformity also in the material considered. It is necessary to bear in mind that, in the reality of the cases, the environments are of scalene dimensions and that the materials used are composites and mixed-layers (Jalali et al., 2008).In order to render the example more realistic a simulation was carried out considering the case of an environment with unequal dimensions on three sides (4 x 5 x 3 $m^3$), with different horizontal wall compositions (masonry- full and perforated ricks) and lower and upper floors (granitic flooring, standard slab).

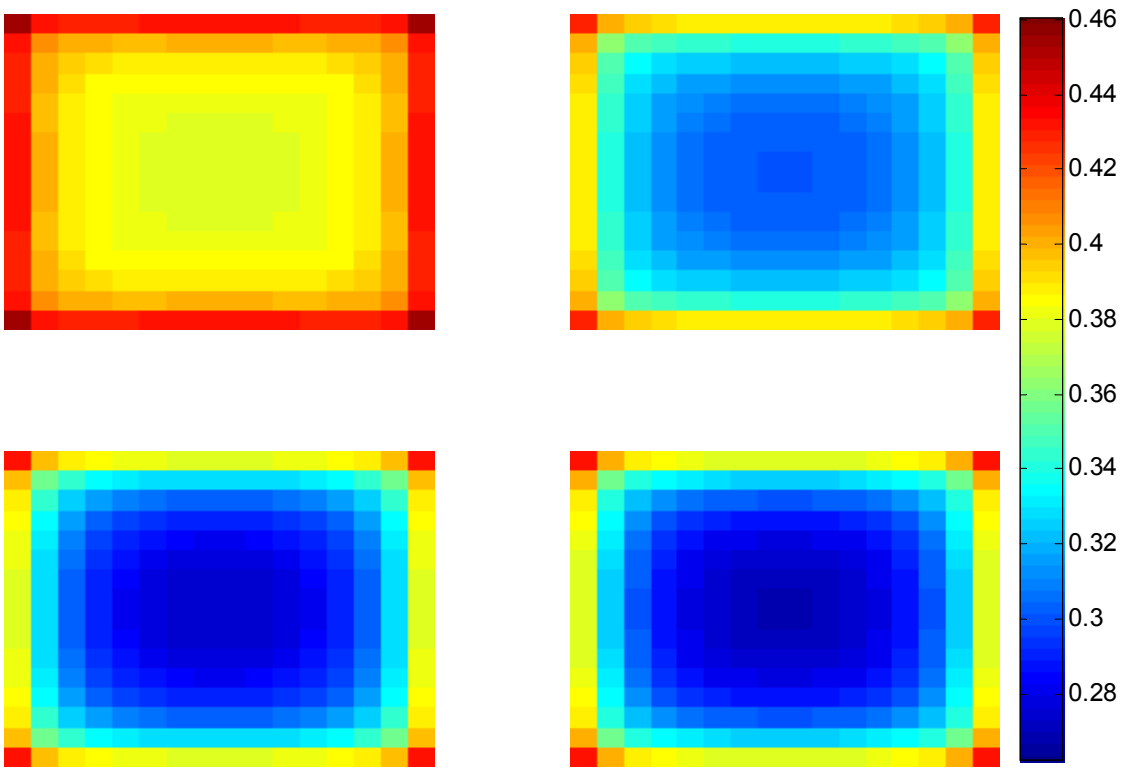


**Fig. 6. Comparative evaluation of the field of radiation in the calculation planes which start from the area adjacent to the wall to the central one of the indoor environment.**

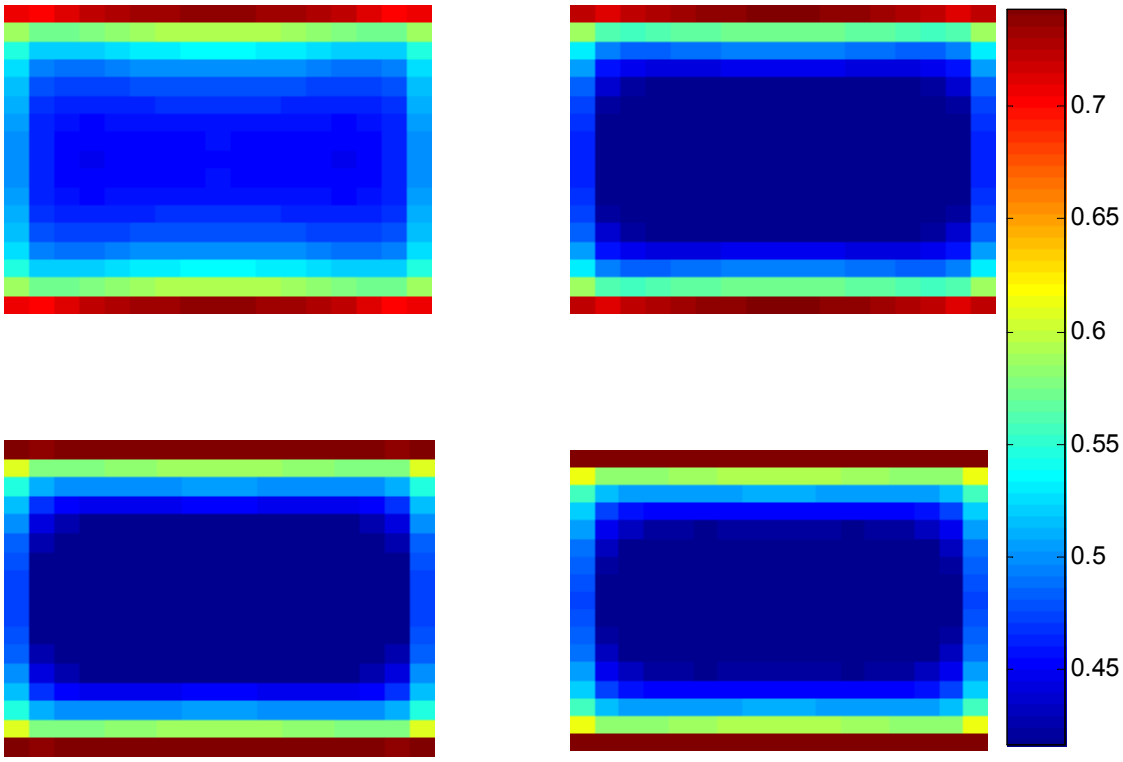


**Fig. 7. Comparative evaluation of the field of radiation for an environment with scalene dimensions with a diversity of materials forming the walls (granite, masonry-bricks).**

Such complications only introduce new weighting factors for the radiation field attenuation, modifying the geometry and symmetry of the field already seen in the previous case.

This new example cannot be exhaustive of the real conditions of use, and therefore of exposure that, obviously, depend on the typical occupation times but also furnishings, the presence of obstacles which more or less influence the trend of radiation.

The following values of concentration of activity referring to the single components of the simulation were inserted:

**Table 2. Activity for materials used for the simulation.**

| Data in literature on commonly used materials (R.P.112) | Average activity (Bq/kg) | | | Conventional dose values (R.P.112 | |
|---|---|---|---|---|---|
| | $^{226}$Ra | $^{232}$Th | $^{40}$K | I | E (mSv/a) |
| **Cement** | 40 | 30 | 400 | 0,42 | 0,22 |
| **Pink Granite** | 150 | 120 | 1600 | 1,63 | 2,46 |
| **Perforated bricks** | 125 | 85 | 1220 | 1,25 | 1,50 |
| **Stucco – plaster** | 10 | 30 | 200 | 0,25 | 0,10 |
| **Wall paint** | 5 | 5 | 15 | 0,05 | 0,01 |

The value of the fluence rate per unit of mass photon flow, in the environment considered in figure 7, has an average value equal to about 0,5. The effective annual dose that results by applying the simulation, results as being equal to E[4] = 1.53 mSv. Such a value corresponds to the corresponding gamma radiation produced by the excess materials as well as the natural dose. This value can be significant of an uncontrolled situation since it concerns the values of indoor exposure to only gamma radiation. The limit introduced by National legislation, which regards exposed workers, imposed an annual limit value equal to 1 mSv. According to epidemiological studies (UNSCEAR 2006), this value corresponds to a probable mortality rate of 0.01%. This value risks further complication due to the coexistence of radon.

The evaluation was made taking in account conversion coefficient in UNSCEAR 1993, 2006 and conversion coefficient from the dose in the air to the effective dose received by an adult individual, 0.8 for the indoor occupation factor, and 0.2 for outdoor (Mulligan et al., 2004). From these values some dose evaluation samples follow, evidently starting from dosimetrical valuations:

Indoors: 84 nGy $h^{-1}$ 8,760 h × 0.8 × 0.7 Sv $Gy^{-1}$ = 0.41 mSv
Outdoors: 59 nGy $h^{-1}$ × 8,760 h × 0.2 × 0.7 Sv $Gy^{-1}$ = 0.07 mSv

The global average resultant for the effective dose is 0.48 mSv, with oscillating values from country to country ranging from 0.3 to 0.6 mSv. For children and infants, the values result as being from 10% to 30% higher, in direct proportion to the increase in the air dose conversion coefficient to the effective dose.

[4] In the dose value nuclides that are more significant to gamma emission, such as $^{40}$K, $^{232}$Th, $^{226}$Ra are taken into consideration.

## Application case study and conclusion

In this last part the possibility of identifying known radioactive sources in structures to be demolished in an indirect way will be shown, examining the model predisposed in the second paragraph.

In the case in which a nuclear plant is being dealt with, for which the principal radionuclides deriving from processes are known, the evaluation carried out previously is not applicable in a direct manner. In fact, considering a uniform distribution of the contaminant agent would not be correct. However, theoretically the considered parameter, Sv, should be substituted with a function that quantifies more or less realistically the contamination situation. In that case it could be possible to adopt a concentration distribution of nuclides equal to an expression such as:

$$Sv = Sv_0 \, e^{-k\left((x-x_o)^2+(y-y_o)^2+(z-z_o)^2\right)} \qquad (11)$$

Where the mass photon flow is defined with $Sv_0$ per volume unit which then is configured in the central contamination position ($x_0$, $y_0$, $z_0$), k represents a scale factor.

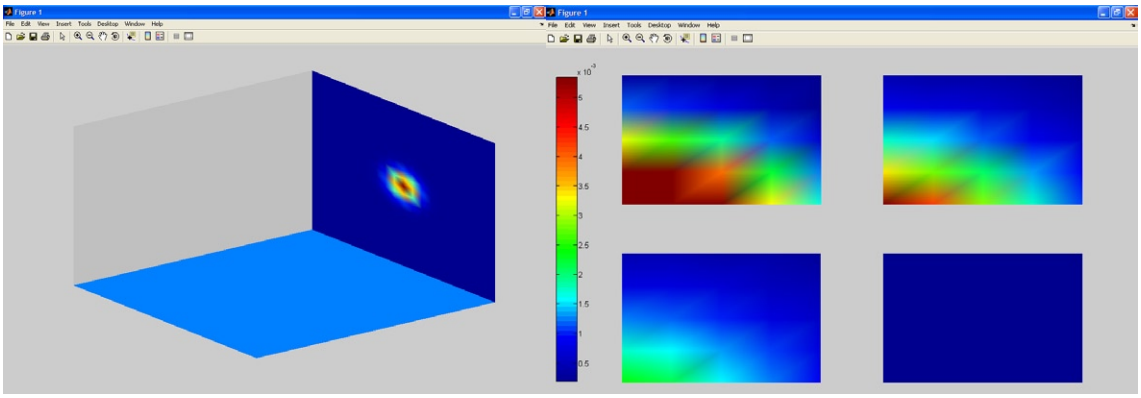

**Fig. 8. Widespread Distribution hypothesis of a contaminated source or NORM (left) and distribution of the field of radiation of a source, concentrated in the corner of a standard room, starting from the wall adjacent to the radiation.**

Within contaminated environments, there are automatic devices which capture the actual activity level (gamma-environment systems, dosimeters, instantaneous detectors, etc.). For practical reasons, such instruments are placed in correspondence of one of the delimiting walls of the environment.

Considering the case of a source that is concentrated in one point of the environment (e.g. in the corner) the distribution of the field of radiation, starting from the calculation plane corresponding to the wall adjacent to the contamination, to the one corresponding to the most distant one (hypothesising a regular environment), is described in figure 8.

Prior to resolving the problem it is first necessary to attempt to formulate it, the problem consists of the evaluation of the entity of a contaminating component, starting from the information provided by in-situ spectrometric data.

As already stated, there is in existence a diversified typology of portable or fixed instruments, even remotely connected, which acquire data providing the instantaneous value of parameters which can be the dose, the activity of a source, the fluence rate etc. Such instruments provide data which is more or less precise of the parameters considered.

Case histories require that the involved radionuclide are known, and with well known emission properties. It all rests upon the determination of the most suitable calculation positions for the evaluation of the actual contamination of the environments: an object of fundamental importance for the entire decontamination process.

The limit of decontamination at the desired clearance level consists, inevitably, of the computation and separation of the natural radiation component, which, as we have already seen, is not negligible and, often, complicates the foreseen removal process.

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